Paulo Alexandre Valente de Jesus Rosa

Mestre em Engenharia do Ambiente

Minimal Computation Structures for Visual Information Applications based on Printed Electronics

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Orientador: António da Nóbrega de Sousa da Câmara, Professor Catedrático, Faculdade de Ciências e Tecnologia da Universidade Nova de Lisboa

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Paulo Alexandre Valente de Jesus Rosa

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Supervisor:

António da Nóbrega de Sousa da Câmara, Cathedratic Professor

Faculty of Sciences and Technology of the New University of Lisbon

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Abstract

In the early nineties, Mark Weiser wrote a series of seminal papers that introduced the concept of Ubiquitous Computing. According to Weiser, computers require too much attention from the user, drawing his focus from the tasks at hand. Instead of being the centre of attention, computers should be so natural that they would vanish into the human environment. Computers become not only truly pervasive but also effectively invisible and unobtrusive to the user. This requires not only for smaller, cheaper and low power consumption computers, but also for equally convenient display solutions that can be harmoniously integrated into our surroundings. With the advent of Printed Electronics, new ways to link the physical and the digital worlds became available. By combining common printing techniques such as inkjet printing with electro-optical functional inks, it is starting to be possible not only to mass-produce extremely thin, flexible and cost effective electronic circuits but also to introduce electronic functionalities into products where it was previously unavailable. Indeed, Printed Electronics is enabling the creation of novel sensing and display elements for interactive devices, free of form factor. At the same time, the rise in the availability and affordability of digital fabrication technologies, namely of 3D printers, to the average consumer is fostering a new industrial (digital) revolution and the democratisation of innovation. Nowadays, end-users are already able to custom design and manufacture on demand their own physical products, according to their own needs. In the future, they will be able to fabricate interactive digital devices with user-specific form and functionality from the comfort of their homes.

This thesis explores how task-specific, low computation, interactive devices capable of presenting dynamic visual information can be created using Printed Electronics technologies, whilst following an approach based on the ideals behind Personal Fabrication. Focus is given on the use of printed electrochromic displays as a medium for delivering dynamic digital information. According to the architecture of the displays, several approaches are highlighted and categorised. Furthermore, a pictorial computation model based on extended cellular automata principles is used

to programme dynamic simulation models into matrix-based electrochromic displays. Envisaged applications include the modelling of physical, chemical, biological, and environmental phenomena.

Resumo

No início dos anos noventa, Mark Weiser escreveu uma série de artigos que introduziram o conceito de *Ubiquitous Computing*. De acordo com Weiser, os computadores requerem demasiada atenção do utilizador, desviando o seu foco das tarefas em mão. Em vez de serem o centro das atenções, os computadores deveriam ser naturais ao ponto de desaparecerem no meio humano. Passamos então a ter computadores verdadeiramente omnipresentes mas também completamente discretos e intuitivos. Para tal, é necessário ter computadores mais pequenos, mais baratos e com consumos de energia menores, mas também soluções de ecrãs igualmente convenientes que possam ser integradas harmoniosamente no nosso meio. Com o recente aparecimento da *Eletrónica Impressa*, novas formas de ligar o meio físico e o digital ficaram disponíveis. Ao se combinar técnicas de impressão comuns tais como a impressão a jacto de tinta com tintas funcionais eletro-ópticas, passou a ser possível produzir em grande escala circuitos eletrónicos extremamente finos, flexíveis e de baixo custo, assim como introduzir funcionalidades electrónicas em produtos onde até agora não era possível devido. Sem dúvida, a eletrónica impressa veio permitir a criação de elementos inovativos para dispositivos interativos, livres de um formato predefinido. Ao mesmo tempo, o aumento da disponibilidade e acessibilidade das tecnologias de fabrico digital, em particular de impressoras 3D, ao consumidor médio está a promover uma nova revolução industrial (digital) e a incitar a democratização da inovação. Actualmente, qualquer pessoa já pode projectar os seus próprios produtos físicos, de acordo com suas necessidades, e fabrica-los sob pedido. No futuro, será o fabrico de dispositivos digitais interativos com formato e funcionalidade específicas que será possível de ser feito a partir do conforto das nossas casas.

Esta tese explora como dispositivos interativos de baixa computação capazes de apresentar informação visual dinâmica podem ser criados utilizando tecnologias de eletrônica impressa e adoptando uma abordagem baseada nos princípios do fabrico pessoal. É dado foco ao uso de ecrãs electrocrómicos impressos como o

meio de transmissão de informação digital dinâmica. De acordo com a arquitectura dos ecrãs, várias abordagens são destacadas e categorizadas. Um modelo de computação pictórica baseada nos princípios dos autómatos celulares é posteriormente utilizado para programar modelos de simulação dinâmicos em ecrãs electrocrómicos matriciais. As aplicações previstas incluem a modelação de fenómenos físicos, químicos, biológicos e ambientais.

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1 Introduction

"The most profound technologies are those that disappear.
They weave themselves into the fabric of everyday life
until they are indistinguishable from it."
Mark Weiser (1991: 1)

1.1 The world is the next interface

If we carefully look around us, it is possible to perceive how computers[1] have become an integral part of our live. They have profoundly and irrevocably changed the way we perform most of our daily tasks, including the way we work, shop, bank, and communicate with our friends and relatives. Simple tasks such as writing a letter, listening to music or reading the news have been utterly altered by computers to a point where most of us cannot imagine realising them without the aid of one. The continuous miniaturisation of microprocessors as well as of other electronic components drove this reality. As computers became smaller, cheaper, and more powerful, their number and presence in our daily life grew remarkably. Moreover, we witness an almost explosive diversification in the nature of computer devices. Nowadays, computers can take various forms and sizes, from smartphones and tablet computers to the credit-card sized Raspberry Pi. Furthermore, they are present and are a crucial component of numerous artefacts and devices such as wristwatches, music players, televisions, washing machines, and microwave ovens.

It is foreseen that in a near future computers will not only be an integrant part of every product we buy but they will in fact be embedded within us and into our environment, inevitably occupying our physical world as natural elements (see for instance, (Gershenfeld, 1999; Greenfield, 2006a; Harper et al., 2008; Norman,

[1] The term "computer" is used here in its broad sense, referring likewise to Personal Computers (PCs) or other simpler computers such as microcontrollers.

1999; Weiser, 1991)). Indeed, computers will become part of the very fabric of our lives.

The proliferation of computers into our physical world promises more than the obvious availability of computing infrastructure anywhere, any time. Computers will enhance our human capabilities and our environment, promoting a reality that is more responsive to our needs and expressive to dynamic changes in its environment. Moreover, it implies a new paradigm of user interaction. The essence of this new paradigm lies in transforming computation, until now essentially focused on point-and-click graphical interfaces, into a new type of user experience, where everything is controlled by natural actions based on our daily activities. We will then be in the presence of intelligent environments, where people do not interact directly with computers but instead are engaged by computer devices of all sizes and types, without necessarily being aware of it (Weiser, 1994, 1999). Computers become not only truly pervasive but also effectively invisible and unobtrusive to the user. Achieving this paradigm shift will require tremendous efforts and creativity from researchers. Not only it will be necessary to develop innovative approaches to link the physical and the digital worlds, but it will also be fundamental to transform the way people perceive computers. Furthermore, new ways to provide and present information will also have to be considered. After all, "*The world is the next interface*" (Gershenfeld, 1999: 169).

1.2 What is wrong with the computer?

Computers are complex machines. Possibly the most complex machine humankind has ever created. However, computers complexity is a misleading one. It results drastically from being a general purpose machine. As Norman (1999) strongly agues in his book "*The Invisible Computer*", the biggest problem of today's computer is that it "*tries to be all things to all people*" (1999: 70).

We use computers to perform the most various tasks. However, this is not necessarily a virtue. As much as the computer empowers us, it also enslaves us. Not only have we become dependent on computers, as they have diminished the control we have over our lives. They impose on us, forcing us to adapt our nature to its terms, rules, and interfaces. Furthermore, computers require too much attention from us. Beyond the constant need of our time and patience due to constant updates and optimisations as well as hardware failures and error messages, computers demand our total focus when using them. Instead of being the centre of attention, computers should be invisible and unobtrusive, part of our human environment (Norman, 1999; Weiser, 1991, 1993b, 1994). Moreover, as Gershenfeld (1999) points out, interaction should happen in the context that we, rather the computer, find meaningful.

1.3 Objective: Invisible technology

Technology should enhance in an invisible and unobtrusive way not only our competences and productivity but also our enjoyment of life. By moving to simpler, task-oriented devices, whose interface reflects the difficulty of the task and not the complexity of the underlying technology, it is possible to keep in sight with our needs (Norman, 1999). This ideal implies the perfect integration between computers and the human environment. Hence, instead of a fixed display, keyboard and mouse, the objects around us become the means we use to interact both with the physical and digital worlds. For instance, tables, walls and floors are transformed into interactive surfaces capable of providing us with subtle visual information about our surrounding, along with the means to act upon it. This requires not only for smaller, cheaper and low power consumption computers but also for equally convenient visual information solutions that can be harmoniously integrated into our surroundings.

With the advent of Printed Electronics, new ways to link the physical and the digital worlds became available. By combining common printing techniques, such as offset printing, flexography or inkjet printing, with conductive inks, it became possible not only to mass-produce extremely thin, flexible and cost effective electronic circuits, but also to introduce electronic functionality in products where it was previously unavailable. As so, we are already witnessing the breakthrough of a completely new set of products where the electrical circuits and some circuit component, such as thin film transistors or resistors, are printed directly in almost any type of organic or inorganic substrates, and thereby becoming an integrating part of the material itself (see (Bennett, 2012; Bliss, 2013; Digital Arts Staff, 2012)). The conjugation with electrochromic inks (for example) enabled, in its turn, the possibility to print and embed displays in these objects and products (see for instance, (Cal Poly, 2012; Ynvisible, 2012)). Moreover, Printed Electronics can create new opportunities for Personal Fabrication by giving individuals the possibility to engineer their own embedded digital devices. Instead of relying on mass-market manufacturers and purchase something that someone else thought they wanted, users can fabricate exactly what they want (see, (Anderson, 2012;

Gershenfeld, 1999, 2005)). Indeed, Printed Electronics can play a major role in the democratisation of innovation. According to Hippel (2005), users' ability to innovate new products and services has been improving radically and rapidly as a result of the steadily improving quality of computer software and hardware, improved access to easy-to-use tools and components, and access to richer libraries of modifiable innovations. Nowadays, it is possible for any individual to easily acquire, at reasonable prices, kits that offer basic electronic and mechanical building blocks. Furthermore, physical product prototyping is becoming easier as computer driven 3D printers become more affordable and sophisticated. The idea of users using the concepts behind Printed Electronics for Personal Fabrication has the potential to empower them, by further enhancing their ability to shape the digital and physical spheres according to their specific needs.

The overall objective of this research work is to further contribute to the Ubiquitous Computing vision and the concept of calm technology by introducing and exploring the use of Printed Electronics and Personal Fabrication technologies in this context. In particular, it aims at the development of task-specific, interactive digital devices capable of presenting dynamic visual information seamlessly integrated in our daily objects and surrounding environment, whilst employing the minimal hardware and software resources. To this aim the following research was carried out to resolve some of the challenges that this vision poses:

1) Based on the foreseen potentials of Printed Electronics, focus was given to it as the preferential approach in the creation of the various components of the digital devices, and to the use of printed electrochromic displays as the medium for delivering digital information. Starting from simple, fixed image, electrochromic displays, it was researched how dynamic visual content could be introduced in these particular type of displays. Direct addressing (segmented) and matrix-addressing electrochromic display were developed as well as the necessary hardware required to control these devices. Various architectures and picture elements arrangements were further explored and categorised based on their potential for presenting dynamic visual information.

2) With the aim to make accessible the development of such devices to wider audiences, including to the common individual, special attention was given to the fabrication techniques, which intended to be as simple as possible and based also on the principles behind Personal Fabrication.

3) A pictorial computation model based on cellular automata principles is later introduced as means to extend the visual potential of matrix-based electrochromic displays. In particular, it is implied its use for creating dynamic simulation models into electrochromic displays by means of various pictorial entities. Envisaged applications include the modelling of physical, chemical, biological, and environmental phenomena.

In sum, this research aims to pave the way towards the consolidation of the Ubiquitous Computing vision via the integration of Printed Electronics technologies whilst also pointing to the disruptive and empowerment potentials of Personal Fabrication.

1.4 Roadmap

The remaining chapters of this document outline the motivation, core concepts and building blocks to support this research as well as the main outcomes. Chapter 2 presents the research rationale and literature review. It starts by providing an historical overview of the vision of Ubiquitous Computing, highlighting the core ideas and concepts behind it. An introduction to the topic of Printed Electronics is followed. The main advantages, challenges and foreseen practical applications are described and an extensive characterisation of the main printing technologies currently used in this area is provided. The last section of this chapter is dedicated to the topic of Personal Fabrication. It makes the connection between the fields of Printed Electronics and Personal Fabrication, and how these can be combined to enhance the state of the art of Ubiquitous Computing.

Chapter 3 explores the importance of visual information and discusses the main non-emissive Printed Electronics display technologies available nowadays. It explains the technology behind each type of display as well as their limitations. Based on the outcome of the review of the various Printed Electronics display technologies, electrochromic displays are pointed out as one of the most promising technologies for the creation of simple, low cost, low power, digital devices capable of providing dynamic visual information.

Chapter 4 represents the lion’s share of the research work. It addresses the development of Printed Electronics devices specifically tailored for visual information applications. The first section of this chapter is dedicated to the development of an overall system architecture based on the use of direct addressing (segmented) and matrix addressing electrochromic displays. The various elements that compose an electrochromic display are described as well as the assembly process of the various devices. The second section, in turn, highlights six main approaches for presenting visual information based on the architectures developed, categorising them according with the capability to produce dynamic content and animations.

Chapter 5 tackles how simple computation programmes such as cellular automata can be a solution for creating complex visual patterns in the proposed systems but as well as a means to perform computations with inherent temporal and spatial dimensions. In particular, it is explored how pictorial entities can be used in a computation model based on extended cellular automata principles to programme dynamic simulation models into matrix-based electrochromic displays.

Finally, Chapter 6 states the conclusions of this work and makes suggestions for future research.

2 The Big Picture: Moving Towards Calm Technology

"The bits and the atoms belong together."
Neil Gershenfeld (1999: 31)

2.1 The third era in computing

The relationship between Humans and Computers has continuously evolved over time. According to Weiser and Brown (1998), in the past fifty years of computation, there have been two main trends in this evolution. These technological changes are essentially characterised by altering the role and place of technology in our lives. They are not actually about the technological developments in itself, but more on how they modify the way we live and work. Nonetheless, each era is a source of technological innovation. They have required the re-opening of old assumptions and the re-appropriation of old technologies into new contexts.

The first era refers to the mainframe relationship (Figure 2-1A). In this era, computers were mostly used by specialists behind closed doors and were considered as a scarce resource that had to be negotiated and shared with others. The Human-Computer relationship was of many people sharing a single computer. The second era is that of the personal computer (Figure 2-1B). As computers entered our houses, they became personal, for individual use. The Human-Computer relationship is characterised by its closeness, where each person has his own personal computer. With the proliferation of the internet and the continuous advances witnessed in computer and communication technologies, we began the transition to the third era: the era of Ubiquitous Computing (Figure 2-1C). This third paradigm pushes computers towards its embedded future, where each person will be engaged by numerous computer devices.

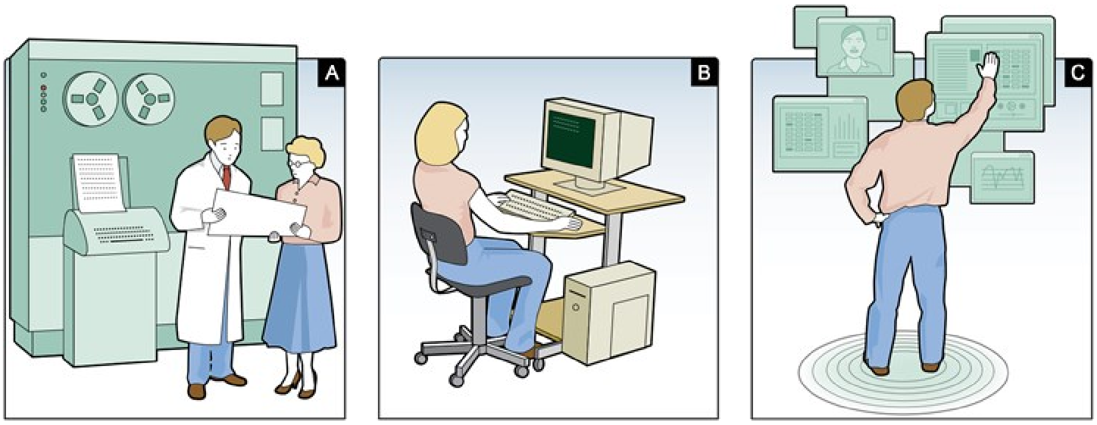

Figure 2-1: The three main eras in computing: A) the mainframe era; B) the personal computer era; C) the Ubiquitous Computing era. Source: adapted from (Harper et al., 2008).

2.1.1 The early days of Ubiquitous Computing: Mark Weiser's vision

Mark Weiser, a chief scientist at the Xerox Palo Alto Research Center (PARC), is widely considered by the scientific community as the father of Ubiquitous Computing, a term he coined in 1988. According to Weiser (1991), the idea of "personal computer" was misplaced and a new way of thinking was necessary. Computers required too much attention from the user, drawing his focus from the tasks at hand. Instead of being the centre of attention, computers should be so natural that they would vanish into the human environment. After all, only when we became unaware of things we are able to freely use them without thinking and therefore fully able to focus in our goals (Weiser, 1991, 1994). Within this vision, computers and others digital technologies are integrated seamlessly into everyday objects and activities, hidden from our senses whenever not used or needed. Technology becomes calm... and its use unconscious.

Calm technology (see (Weiser and Brown, 1995, 1998)) engages both the centre and the periphery of our attention, moving back and forth between the two whenever necessary. It portrays a world of serenity, comfort and awareness, where we are continuously kept informed of what is happening in our surroundings as well as what has just happened and what is going to happen without being overburdened. Information would appear in the centre of our attention when needed and effortlessly disappear into the periphery of our attention when not. What is in the periphery at a given moment can move to the centre in the next one, becoming the focus of the attention. Weiser and Brown (1995) argue that this is fundamentally "*encalming*" for two reasons. First, by placing things in the periphery we became capable to attune many more things than we could if everything had to be at the centre. We are unconsciously aware of what is happening around us, what has just happened and what is going to happen. The periphery functions as a mean of collecting information without distracting or overburdening. Second, by re-centring something formerly in the periphery we take control of it. Peripherally we become aware of things and by centring them we enhance our awareness and power regarding that specific item or event.

Another important facet of Weiser’s vision is the ability of each single computational device to interact with the nearby ones. They will all be wirelessly interconnected and each user will be able to interact with several computational devices simultaneously without necessarily realizing it. Information will move from one device to another seamlessly and will be accessible to users anywhere, anytime. Furthermore, computers will be aware of their location. They will be able to adapt their behaviour in significant ways without requiring even a hint of artificial intelligence (Weiser, 1991).

Box 2-1: Early Ubiquitous Computing developments.

Tabs, Pads and Boards

Weiser (1991, 1993a) predicted that ubiquitous computers would come in different sizes, each tailored for a particular task. Having this in mind, his team at Xerox PARC developed a variety of Ubiquitous Computing prototypes, namely a series of Tabs (Want et al., 1995), Pads (Kantarjiev et al., 1993) and Boards (Elrod et al., 1992), along with the necessary infrastructures which allowed these devices to talk among themselves.

Tabs (shown in Figure 2-2) were the smallest of the three devices and were designed to mimic the concept of active post-it notes. Each tabs had a pressure sensitive screen on top of the display, three buttons underneath the natural finger positions and the ability to sense its position within a building. They had very limited processing capabilities but also very low power consumptions.

Pads were similar, in size and in intended behaviour, to a sheet of paper or a book. They were developed to be used as “scrap computers”. They had no individualised identity or importance. Instead of being carried from place to place by a specific owner, they were to be used and left somewhere for someone else to use them again.

Boards were wall-sized devices with huge interaction areas. They were the equivalent of a blackboard or a bulletin board. In a home environment, they could be used as video screens while at work they function as whiteboards or flip charts. The information within one board was easily shared among other boards, even if they were located in different rooms.

Figure 2-2: Picture of a Xerox PARC tab. Source: (Greenfield, 2006b).

The true power of Ubiquitous Computing comes not from any of these devices in itself but emerges from the interaction of all of them. "*For each person in an office, there should be hundreds of tabs, tens of pads, and one or two boards*" (Weiser, 1993a: 76).

Unfortunately, these initial prototypes were not capable to successfully implement Weiser's idea. They failed to become invisible, and furthermore, they did not manage to convey the interface vision well, ending up inheriting all the interaction problems typically present in a graphical user interface driven device.

Active Badge Location System

The "Active Badge Location System", developed by Want *et al.* (1992) is one of the earliest and more widespread examples of a Ubiquitous Computing prototype development. The system allowed the automatic location of individuals within a building by determining the location of their Active Badge (Figure 2-3). Each badge transmitted a unique infrared signal that was detected by a network of sensors equipping the building.

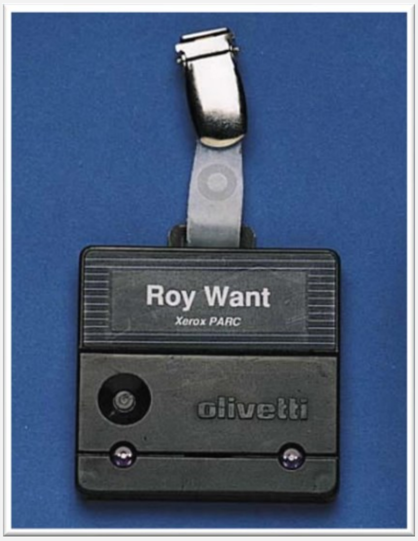

Figure 2-3: Picture of an Active Badge. Source: (AT&T Laboratories Cambridge, 2002).

Further versions of the Active Badge were developed in order to expand its functionalities. In a more recent version, the badge incorporated a small display than could serve simultaneously as an active badge, a calendar and a diary. Individuals wearing this badge could automatically unlock restricted areas to which they had been granted access, have phone calls routed to them wherever they were, and create diaries of meetings they had attended. This implementation clearly illustrated how a simple identity tag could be transformed into a multi-functional platform.

The implementation of the Active Badge Location System, nonetheless, raised many concerns regarding privacy issues. Since it allowed the location of any individual within the working area, the fear that the system could be used to spy workers was always present.

2.1.2 Ubiquitous Computing nowadays

Countless efforts have been put forward in transforming Weiser's Vision into reality. Human environments have been augmented with diverse computational devices that enable people to engage and access information and services when and wherever they desired. Sensors have been implemented not only in our houses and offices but also in our bodies providing overwhelming amounts of data about our environment, our movements and our health. Furthermore, these data have been increasingly used to automate mundane operations and actions that we do in our daily life using conventional physical controls such as buttons and switches (Rogers, 2006).

It is already possible to point out numerous devices that are clear indicators of this new technological revolution. Mobile phones and smartphones with internet capabilities, electronic labels and RFID (radio-frequency identification) tags, miniature cameras and flexible displays, are just some examples of the technologies currently available that are driving us towards the "post-PC era". In fact, devices such as the e-book reader and the tablet computer are roughly overcoming the paradigm of the general-purpose personal computer in favour of simple, specialised digital devices integrated in our life style. As Mattern (2004) points out, the technological bases for a new world are already here.

It becomes also evident that the idea behind Ubiquitous Computing became broaden. Weiser initial concept was further explored by other scholars (see for instance, (Greenfield, 2006a; Mattern, 2001; Sakamura, 1996)) and rephrased to incorporate new domains of application and novel concepts. The terms Pervasive Computing, Mobile Computing and Ambient Intelligence became a synonym of Ubiquitous Computing and expressions such as physical computing, smart devices, tangible media or the Internet of Things are commonly used to describe the devices involved to power these concepts (see (International Telecommunications Union, 2005; Mattern and Sturm, 2003; Ullmer and Ishii, 2001)). While a number of authors defend that each concept is in its core different, others point out that the differences between these terms is merely of academic nature. For example, Lyytinen and Yoo (2002) argue that the concepts of Mobile Computing and Pervasive Computing, despite being often used as synonyms of Ubiquitous Computing, are conceptually different and employ different ideas of organising and managing computing services. While Mobile Computing is fundamentally about improving the capability to physically move computing services between environments, Pervasive Computing refers to the capability that computers have to obtain the information from the environment in which they are embedded and to use that information to dynamically built computational models. Hence, the authors present Ubiquitous Computing as the integration of large-scale Mobile Computing with the Pervasive Computing functionality. Greenfield (2006a), on the other hand, considers all these concepts facets of one coherent paradigm of interaction that he prefers to calls "everyware". He points out that there are many forms of Ubiquitous Computing, and regardless of the different and valid distinctions between each definition, they are all indistinguishable and meaningless from the user's perspective. Hence, to the user, they are all aspects of a single paradigm.

The multiple variations of concepts can be seen as a product of the natural evolution of technology. As technology evolved, the new discoveries pushed the limits of the existing definition. In order to accommodate the new knowledge and expertise, the concept of Ubiquitous Computing, inevitably become broader. Jeon et al. (2007) argue that the important thing is to redefine the different concepts as a general notion that incorporates the basic ideas suggested by all of them. The authors propose a taxonomy of properties from which the different kinds of ubiquitous systems and applications can be described and compared. "*In order to understand Ubiquitous Computing environment, overall characteristics of Ubiquitous Computing should be considered first*" (Jeon et al., 2007: 1230).

Independently of the various uses given to the concept of Ubiquitous Computing and of the different terminologies adopted, the following basic principles are always present:

- Ubiquitous Computing devices are integrated seamless into everyday objects, becoming invisible. Interaction becomes natural, unconscious.

- Ubiquitous Computing devices are linked by wireless networks, becoming available anywhere, anytime.

- Ubiquitous Computing devices are able to communicate among them and to adapt to variations in the surrounding environment.

However, Mark Weiser's vision of Ubiquitous Computing was not primarily about the idea of "anytime and anyplace" computing. In fact, when Weiser coined the term Ubiquitous Computing, this notion was already employed in a variety of contexts. Nonetheless, as Ishii (2004) points out, Ubiquitous Computing ended up being used as a new label for an old idea. Weiser´s concept was misunderstood and, as a consequence, usually misapplied (see Weiser comments in (Ishii, 2004)). Weiser´s idea was centred on the context of interface design. It "*was never just about "making" computers ubiquitous. It was always* […] *about awakening computation mediation into the environment*" (Weiser in Ishii, 2004: 1310). As a way to stress and reinforce the ambient interface aspect, Weiser introduces the concept of Calm Technology. However, the term Ubiquitous Computing and its misleading connotations were already too popular in the field of computer science. The effect obtained in influencing and shifting the current research was minimal.

Ishii and Ullmer (1997) stimulated by Weiser vision of Calm Technology, developed the concept of Tangible Bits. Like Weiser, they pursued the vision of moving beyond the traditional point-and-click graphical interfaces, to one that disappears into everyday objects. They explored novel ways of re-applying augmented physical objects commonly used in daily life with digital technology. Their aim was to bridge the gap between the digital and the physical environment by making digital information tangible.

Box 2-2: Examples of Calm Technology.

ambientROOM

The *ambienteROOM* (Ishii et al., 1998) is a personal interface environment designed to engage both the centre and the periphery of the user attention. It consisted of an office room where several areas of the space were augmented to display and communicate information through subtle cues of sound, light, or motion. In addition, graspable objects, such as bottles, were used as a mean to control the ambient media. The *ambienteROOM* environment explored the periphery of human perception in computational environments through the implementation of novel interfaces that merged the digital and the physical environments.

Bottles

Bottles (Ishii, 2004; Ishii et al., 2001) is a multi-modal user interface that uses glass bottles as "containers" and "controls" of digital information. In one of the first projects, *musicBottles* (Ishii et al., 1999), each bottle had a different music instrument associated to it, behaving like if it was "filled" with music. Opening a bottle, by removing its cork, would release the sound of the music instrument from its inside. A piece of music would then begin to play, accompanied by a dynamic coloured light. The physical manipulation of the bottles, opening and closing them, was the primary mode of interaction for controlling their musical contents. By integrating glass bottles, a custom designed table, music, and colourful lighting, the authors hoped to create an engaging and aesthetic interface that could provide rich emotional experiences to users who were unfamiliar or uncomfortable with current personal computers.

Figure 2-4: *musicBottles* installation. Source: (Ishii et al., 1999).

In contrast, Rogers (2006) argued that Weiser's vision of Calm Technology gives the user a very passive role, leaving him in a quite idle position as just the receiver of information. The author defends that "*we need to rethink the value and role of calm and proactive computing as main driving forces*" (Rogers, 2006: 406). He proposes an alternative approach in which the focus is on designing Ubiquitous Computing technologies for engaging user experiences. Instead of computers, people should take the initiative to be constructive, creative and ultimately to control their interactions with the world. "*Rather than calm living it promotes engaged living, where technology is designed to enable people to do what they want, need or never even considered before by acting in and upon the environment*" (Rogers, 2006: 406). Nonetheless, as the author points out, this does not mean that the main principle of Weiser's vision should be discarded. Simply, there are also other valid possibilities that can be considered for steering Ubiquitous Computing research.

It is also comprehensible that the shift towards Ubiquitous Computing poses multiple challenges, both at a technical and social level (Bohn et al., 2005; Chalmers et al., 2006; Harper et al., 2008; Kallio and Latvakoski, 2004). As Weiser clearly points out, "*getting the computer out of the way is not easy*" (Weiser, 1993a: 76). It requires not only a clear transformation of the context of usage of the machine and of its physical elements as well as the creation of a new kind of relationship between the user and the computer. Chalmers et al. (2006) argues that it is fundamental to explore novel techniques that support interaction with and through new types of computational devices. Gesture-based approaches exploiting movement in relation to surfaces and artefacts; haptic approaches exploiting the physical manipulation of artefacts; and speech-based interfaces; need to be further studied and developed. In addition, these solutions must fit the user's needs, goals and skills.

New ways to provide and present information, both visually and non-visually, also need to be envisaged. Users must be able to easily access the information in a comprehensive and clear way. Moreover, in order to effectively design systems that can be perceived both in the periphery as well as in the centre of the attention, a detailed understanding of not only how information can be presented but as well as how it is perceived at the different levels of the human attention must be procured. Naturally, it becomes also important to consider how these transitions between the different levels of awareness can be eased and smoothed for the user experience (Bakker et al., 2010; Brown and Duguid, 1996; Eggen and Mensvoort, 2009).

Understanding how users will interact and experience Ubiquitous Computing technologies and how these can be integrated in human activities along with its consequences, is fundamental for the creation of useful and suitable interfaces and interactions. Ubiquitous Computing systems must be sufficiently simple and transparent so that people can understand effortlessly how to interact with them. Hence, it is necessary to develop all-inclusive standardised invisible interfaces. New hardware components have to be assembled, new network protocols created and software developed but also further studies have to be conducted to better understand the interaction between Man and Machine. The social and cultural realms also pose a great importance and must also be taken into consideration.

Always present are as well concerns about invasion of privacy; data and consumer protection; trust, accountability and security of systems; and loss of control (Friedewald and Raabe, 2011; Hayat et al., 2007; Want et al., 2002). Naturally, users will want to take advantage of the Ubiquitous Computing potential and be able to engage and be engaged by every object they encounter without worrying about security and privacy issues. Success in addressing these challenges will inevitably require the expansion of Ubiquitous Computing research to areas outside computer science. Hence, as mentioned above, it becomes fundamental to transpose the traditional barriers between the social and the technical realms and promote the close collaboration between computer engineers and social theorists. Indeed, given the potential changes that Ubiquitous Computing can bring to business practices, commerce, governance, and overall everyday life, there is huge potential for social science research.

2.2 The advent of Printed Electronics

When Johannes Gutenberg invented the movable type printing press around 1440, he initiated a revolution in the distribution of knowledge. His invention allowed that individual letters and punctuation symbols could be used over and over again to print the words and sentences in each page of a book, making it possible to produce a large number of copies of a single work in a relatively short amount of time, considering the previous alternative of manual copying. By the end of the fifteenth century, hundreds of book titles were being produced each year on wooden presses similar to the one developed by Gutenberg. This rapid spread of knowledge made possible by Gutenberg's printing press ended up having a fundamental role in the development of the Renaissance, the Protestant Reformation and the Scientific Revolution (Gutenberg Museum Mainz, 2008; Harry Ransom Center, 2007). In an analogous way, Printed Electronics intends to revolutionise the production of digital devices by combining the achievements of the printing industry and those of the electronics world. The term Printed Electronics refers to the use of printing technologies to produce electronic circuits, components and devices in a wide array of substrates, such as paper, plastic or textiles. Electro-optical functional inks are used for this purpose, which are directly deposited on the substrate, creating the various active and passive devices (e.g. transistors, resistors, capacitors, antennas, and alike).

The interest in Printed Electronics lies primarily in the prospect that printing is a low cost technique for the production of electronic systems, ultimately capable of bringing down the manufacture cost of such products to values that conventional silicon manufacturing cannot reach. The potential for cost savings comes from the fact that Printed Electronics is based on the use of purely additive processing methods, in contrast to the photolithography-based subtractive methods currently used in the semiconductor industry (Subramanian et al., 2008). Not only is the material only deposited where it is required, but also the overall complexity of the manufacture process is greatly simplified. Typically, only two steps are required to go from a bare substrate to a working functional layer on a substrate: the printing process in itself and a curing process. If we consider that in subtractive methods multiple steps, materials and equipments are necessary to produce a single functional layer on a bare substrate, in addition to being consumed materials that do not end up on the final device, the cost savings can be relatively high, particularly when the device does not have a high surface coverage on the substrate (Ghaffarzadeh, 2013). In sum, Printed Electronics has the potential to simplify the process flow, increase the material utilisation, shorten the value chain, and decrease the overall tooling cost, therefore reducing capital expenditure and increasing throughput across the entire flow (Ghaffarzadeh, 2013; Subramanian et al., 2008).

However, there is a trade-off. Printed Electronics components do not have the same high performance and reliability as their non-printed counterparts (Subramanian et al., 2008; Volkman et al., 2004). Hence, it is not expected that Printed Electronics will substitute conventional silicon-based electronics, at least in a near future. Instead, it can be seen as an entirely new market and industry. Printed Electronics represents a ground-breaking new type of electronics that are as well characterised for being lightweight, thin, flexible, robust, and easily disposable. It opens a new world of opportunities for low-cost printed circuits aimed at high-volume market segments where the high performance of conventional electronics is not required (Kantola et al., 2009) as well as for low level prototyping. Moreover, it enables a new set of opportunities and possibilities for products and applications by allowing the incorporation of electronic functionalities into artefacts where it was previously unavailable. Indeed, Printed Electronics can become a mean for transforming lifeless objects and surfaces into sensing, interacting interfaces, capable of reacting and exchanging information with users and the environment.

Table 2-1: Comparison between Printed Electronics and conventional electronics.

	Printed Electronics	Conventional Electronics
Performance	Low	High
Area per Feature Size	Large	Small
Cost per Unit Area	Low	High
Throughput	High	Low
Substrate	Flexible	Rigid
Product Lifetime	Short	Long

At present time, the market drivers for Printed Electronics are radio frequency identification (RFID) tags (Chan et al., 2005; Subramanian et al., 2005, 2006; Yang and Tentzeris, 2007); memory (Allen et al., 2011; Andersson et al., 2011; Kim et al., 2011; Lian et al., 2010) and logic components, including field effect transistors

(FETs) (Hartïng et al., 2009; Schneider et al., 2008) and thin film transistors (TFTs) (Burns et al., 2002; Kawase et al., 2003; D Kim et al., 2009); sensor arrays (Harrey et al., 2002; Honeychurch and Hart, 2003; Laschi et al., 2006; Li et al., 2007); photovoltaic cells (Barr et al., 2011; Krebs, 2009); batteries (Blue Spark, 2011; Enfucell, 2012; Hahn and Reichl, 1999; Hilder et al., 2009); and displays (see section 3.2). The practical applications envisaged are various, and include, for example:

- Dynamic newspapers, magazines, and signage applications: By taking advantage of the combined benefits of paper with dynamic digital content, companies can create novel formats to present visual information and publicise their products. This will likely include the incorporation of animated advertisements in magazines and newspapers, or the creation of dynamic signage and billboards. Other possibilities include, but are not limited to, posters, business cards, bumper stickers, and product packages and labels.

- Intelligent packaging: Printed displays can be incorporated into products packages not only with the aim of making them more visual appealing and attractive, but also more useful and helpful. Hence, Printed Electronics systems can be used in packaging to improving the legibility and detail of the information available about the product, and thus improving the information that consumers have access in the act of purchase, or can be used to show notice messages about the conditions of the product, highlighting changes that occurred in the surround environment and that are incompatible with the preservation of the product. Furthermore, anti-counterfeiting measures can also be implemented directly into the products, preventing or at least complicating the falsification of products.

- Smart labels: From low-cost remotely updated electronic shelf labels and pricing tags used in supermarkets and stores to dynamic information labels for products.

- Smart cards: The implementation of Printed Electronics systems in smart cards could allow users to rapidly access information contained in the card, wherever and whenever they wanted. This would enable, for instance, customers to easily check the amount of credits still remaining in a public transportation smart card, or the validity of their subscription. Frequent flyer card, or in any other type of loyalty system card could indicate the fidelity points gathered, or alert the user for promotions. Healthcare smart cards could also be enhanced, allowing users to easily check certain information on their medical file, such as the blood type, whether the vaccines are up to date, when it was the last time he went to the doctor or when he is supposed to have is next medical visit. Furthermore, Printed Electronics solutions could also be used to improve the security of smart cards, especially of debit and credit cards (e.g. by implementing digital watermarks).

- Healthcare diagnostic devices: The disruptive potential of Printed Electronics can be enormous in the healthcare sector. By enabling the fabrication of disposable printed biosensors at a fraction of the cost of equivalent non-printed solutions, they can make complex healthcare examinations not only cheaper but also faster to do. These biosensors are traditionally used in medical monitoring, diagnostics, and drug delivery. Examples include biosensors for monitoring vital signs (e.g. heart rate, body temperature, blood pressure); for testing metabolic variations (e.g. blood glucose, cholesterol, lactate); and for detecting pathogens elements (e.g. bacteria and virus).

- Energy harvesting and storage devices: Various printing technologies are already being used as fabrication tools for manufacturing photovoltaic cells and batteries. As printed photovoltaic cells become more efficient and more reliable as a power source, they will eventually become more widespread. Low-cost printed photovoltaic cells will allow energy to be generated where it is needed. Considering their flexible nature, they can be easily integrated into building structures, such as wall coverings, or made into window shades. Likewise, printed batteries provide lightweight, flexible power sources that can be integrated into mobile electronic devices, or in any other type of low-power consumer application or Printed Electronic system.

- Dynamic walls and lighting panels: Printed Electronics systems can be integrated into walls and be used as information screens or, alternatively, as dynamic wallpapers or lighting panels.

- Active/smart clothing: Printed Electronics systems can also be integrated seamlessly into textiles. They can be used to improve the functionality of clothes, for instance, by using embedded biosensors and displays to monitor and show the user vital signs, or instead, in a more fashionable way, to simply display dynamic patterns in the fabric.

Naturally, the development of these applications is greatly conditioned by the formulation of suitable functional inks as well as of adequate substrates where they are printed (Kantola et al., 2009). After all, the practicality of Printed Electronics relies primarily on the development of novel inks used to create the electronic components. As for substrates used, so far the most common ones are polymer films, ceramics, glass and silicon. Printing of functional inks on paper is also possible, but can present some challenges due to the paper's rough, fibrous surface at a microscopic scale. The optimisation of current printing technologies for real mass-manufacturing of Printed Electronic systems also has to be undertaken. As Schmidt *et al.* (2006) points out, printing technologies were developed for visual output and therefore classical printing products undergo completely different requirements when compared to electronic devices. Significant modifications in processes and materials are necessary.

For the purpose of this thesis, a digital device is regarded as an electronic system that can receive, store, process and display digital information. It is composed by the three simple interconnect modules: (1) the sensing module, responsible for picking up the user inputs; (2) the computation module, responsible for the data processing, transforming the inputs into relevant outputs; and (3) the display module, responsible for outputting the results of the computation into a human-readable form. A fourth module, the power module, can also be considered, being responsible for supplying the necessary energy for the smooth operation of the system.

2.2.1 Printing technologies

The assortment of printing technologies currently available for the production of traditional printed products is relatively broad. Based on whether a master, i.e. a printing plate or an image carrier from which the ink is transferred to the printing substrate in order to reproduce text or images, is required or not, printed technologies can be divided into conventional printing and non-impact printing, respectively (Kipphan, 2001).

In conventional printing, the master is the medium responsible for carrying the information during the printing process. As a result, information is generated on the printing substrate based on the master layout, through direct contact and by partial transfer of the printing ink. Each master corresponds to a single page of information. In contrast, in non-impact printing, there is no master carrying permanent information, and text and images are produced without direct physical contact between the printing mechanism and the printing substrate. Hence, different information can be printed continuously on each page. The printing process is based on digital information and, depending on the applied non-impact printing technology the image is either transferred via an intermediate carrier/imaging surface (e.g. electrophotography printing), or is transferred directly without an intermediate carrier (e.g. inkjet printing). An overview of the various printing technologies is provided in Figure 2-5.

Within the context of Printed Electronics, the most commonly used printing technologies are screen, flexography, offset lithography, gravure, and inkjet. Naturally, each process has its own strengths and limitations in regard to the production of Printed Electronics. The choice of one process over another is typically related to the type of ink, substrate used, and the final application intended (for instance, prototyping versus high-precision). Hence, each process tends to be the ideal method of production for a different range of products or substrates. In order to fully take advantage of the production capabilities of conventional printing technologies, their applicability in Printed Electronics should be target to roll-to-roll processing (R2R). This essentially consists of adapting the printing technologies to allow rotary printing. The process typically involves several rotating cylinders around which the printing substrate is routed through a number of fabrication operations. Hence, during the printing process, the substrate is on a constant move and the imprint is done in a continuous process at impressively high speeds, enabling large area capability, high throughput, and ultimately increasing the cost-efficiency of the overall manufacture process.

In the following sub-sections, the most commonly used printing technologies in Printed Electronics are described, highlighting their advantages and limitations. Table 2-2 provides a comparative overview of these technologies.

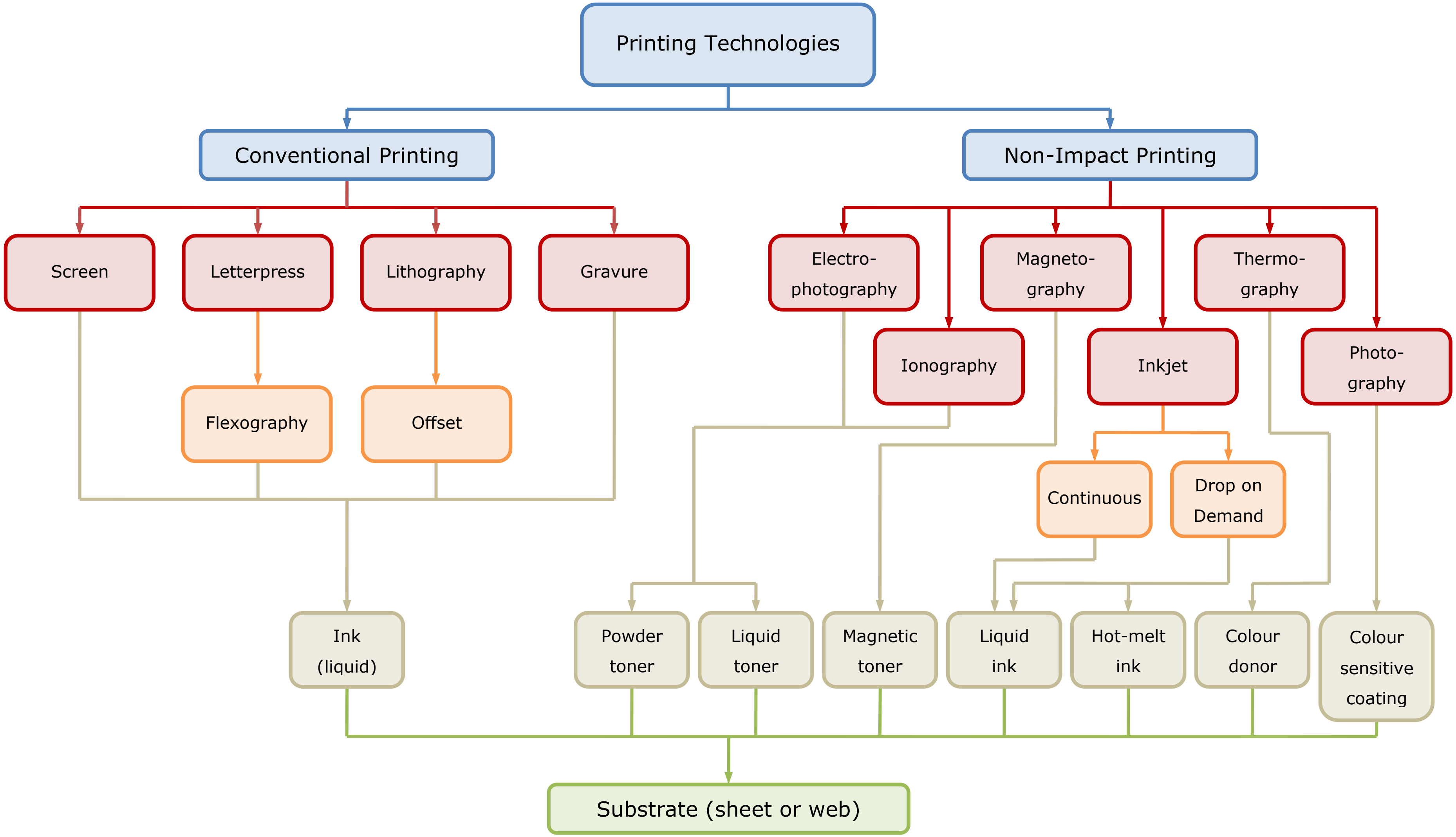

Figure 2-5: Overview of printing technologies. Source: adapted from (Kipphan, 2001).

Table 2-2: Comparison of printing technologies typically used in Printed Electronics.

	Screen Printing	Flexography Printing	Offset Printing	Gravure Printing	Inkjet Printing
Printing Form	Stencil	Relief	Flat	Engraved	Digital
Image Transfer	Direct, wrong reading	Direct, wrong reading	Indirect, right reading	Direct, wrong reading	Direct, non-impact
Resolution (lines/cm)	50	60	100 to 200	100	60 to 250
Line Width (µm)	50 to 150	20 to 50	10 to 15	10 to 50	1 to 20
Ink Viscosity (Pa·s)	> 1 to 50	0.05 to 0.5	40 to 100	0.05 to 0.2	0.001 to 0.03
Film Thickness (µm)	up to 12	1 to 2.5	0.5 to 1.5	<0.1 to 5	0.5 to 15
Printing Speed (m/min)	10 to 15	100 to 500	200 to 800	100 to 1000	15 to 500

Source: (Caglar, 2009; Kipphan, 2001; Romano, 1999).

2.2.1.1 Screen printing

Screen printing is possibly the most mature technique for the fabrication of Printed Electronics. In the last decades, it has been widely applied in the fabrication of printed circuit boards (PCBs), namely to pattern conductor traces, typically using silver pastes; resistors, using carbon films; and capacitors, using polyimide dielectrics (Subramanian et al., 2008). It is a rather inexpensive and highly flexible process for manufacturing electronics.

In its core, screen printing consists of an interlaced mesh screen through which ink is forced to pass so as to imprint an image onto a substrate surface. A stencil, which is coated onto the mesh, is typically used as the carrier of the printed information. Hence, in screen printing, the printing plate is in reality a combination of the mesh and the stencil. The image to be printed is defined by the open areas of the otherwise filled stencil/mesh. A squeegee, i.e. a type of blade, which moves relative to the mesh screen, is used to force the ink through the areas that are not covered by the stencil onto the substrate. The mesh is typically made of a fine fabric of natural silk, plastic, or metal threads, being the last two more common nowadays (Kipphan, 2001). The thread diameter, the mesh count, and the solid content of the ink determine the amount of ink that passes through the mesh. The same image can be reproduced numerous times using the same mesh screen.

Screen printing is one of the most versatile processes for transferring ink onto a substrate. When compared to the other printing technologies, it provides the widest range of application with regard to the choice of substrates. Apart from paper and cardboard, other possible substrates are plastics, glass, metal, textiles, ceramics, and the like, whether in the form of endless webs or single sheets. Moreover, the substrate surface does not need to be planar, and thus objects of the most varying shape can also be used as printing substrate. The range of suitable inks is as well high. However, these need to have a paste-like behaviour. Screen printing requires rather high viscosity inks, typically superior to 1000 cP (1 Pa·s), with thixotropy[2] properties. Inks with lower viscosities simply run through the mesh and cause excessive spreading (Søndergaard et al., 2013; Subramanian et al., 2008). It must be pointed nonetheless, that the use of high viscosity inks raises some issues in the field of Printed Electronics. High viscosity inks are typically manufactured by adding polymer binders to the ink, and these binders can destroy the functionality of semiconductors, introduce excessive leakage and dissipation in dielectrics, or degrade the conductivity of conductors (Subramanian et al., 2008). Another characteristic of screen printing is that a greater thickness of ink (up to 12 µm) can be applied to the substrate than it is possible with any other printing technique (Kipphan, 2001). This allows the patterning of very thick dry films, which can be extremely useful for printed electrodes where high conductivity is needed.

In the context of Printed Electronics, screen printing has been widely used in the production of polymer photovoltaic cells, to print both the front and back electrodes as well as of complete cell modules (see for instance, (Aernouts et al., 2004; Krebs, Gevorgyan, et al., 2009; Krebs, Jørgensen, et al., 2009; Shaheen et al., 2001). Other examples include the production of displays, from electrochromic displays (Brotherston et al., 1999; Coleman et al., 1999; Pettersson et al., 2002) to organic light-emitting diode (OLED) displays (Birnstock et al., 2002; Jabbour et al., 2001; Pardo et al., 2000) and field emission displays (FED) (Yukui et al., 2002; Zeng et al., 2006); RFID antennas (Kim et al., 2012; Shin et al., 2009); and various types of sensors (Hart and Wring, 1997; Hart et al., 2005; Honeychurch and Hart, 2003; Laschi et al., 2006; Patel et al., 2001).

Figure 2-6 illustrates the traditional flatbed screen printing configuration and the continuous rotary screen printing method.

[2] Thixotropy consists of a reversible micro-structural change in the viscosity properties of a element when submitted to shear stress. These elements are characterised by having a high viscosity at low stress levels, but a decreased viscosity when an increased stress is applied (Barnes, 1997).

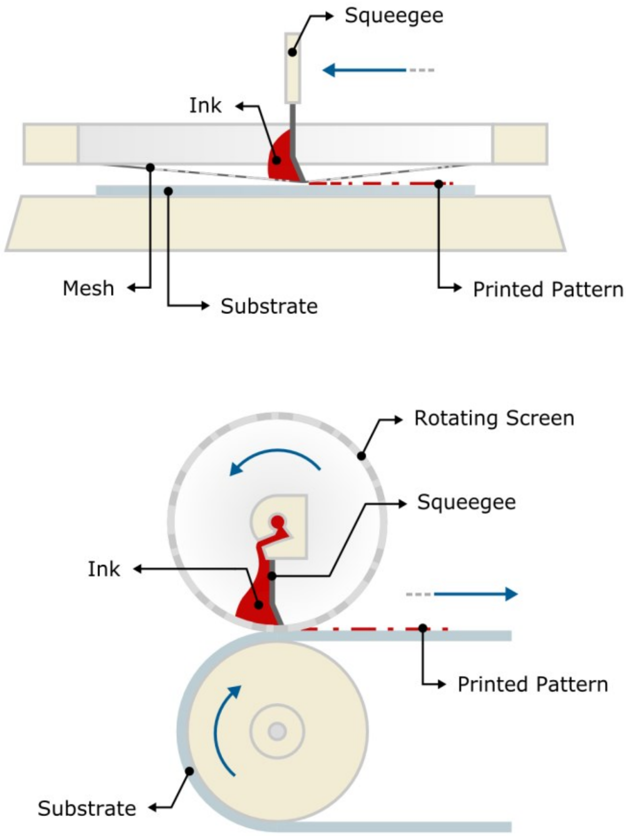

Figure 2-6: Schematic illustration of the flatbed screen printing (top) and rotary screen printing (bottom) methods. Source: adapted from (Kipphan, 2001).

2.2.1.2 Flexography printing

Flexography printing, in the simplest arrangement, consists of four elements: the impression cylinder; the printing plate cylinder; the anilox roller; and the fountain roller. Images are printed onto a substrate by means of a cylindrical printing plate, typically made of rubber or a photosensitive plastic material (i.e. a photopolymer). The pattern to be printed is engraved as a positive relief in the printing plate soft structure, and through direct contact, ink is transferred to the substrate, therefore reproducing the desired pattern. A ceramic anilox roller, with a specifically designed exterior texture formed by engraved microcavities, controls the amount of ink that is delivered to the printing plate cylinder. In turn, the anilox roller is continuously supplied with ink through the so-called fountain roller, which is partly immersed in an ink bath. Any excess of ink in the microcavities of the anilox roller is removed by a type of blade named doctor blade. The thickness of the printed film is defined by the volume of the microcavities in the anilox roller, as well as by the transfer rates from the printing plate cylinder to the printing substrate (Caglar, 2009; Kipphan, 2001; Søndergaard et al., 2013). The basic principle on which flexography printing operates is illustrated in Figure 2-7.

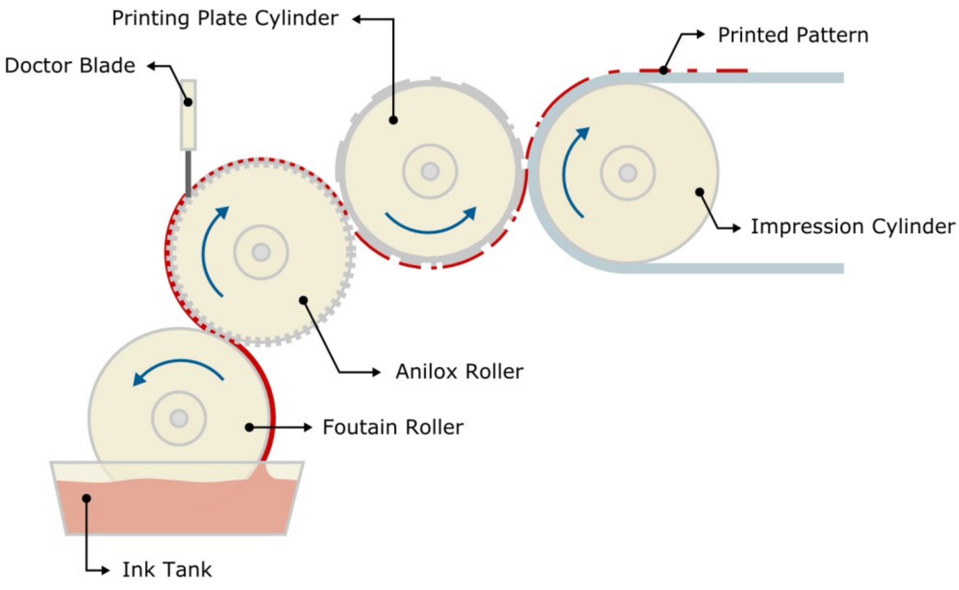

Figure 2-7: Schematic illustration of the flexography printing process. Source: Adapter from (Kipphan, 2001).

Flexography main advantage is its ability to print on a wide variety of materials, allowing printing substrates to be chosen based on their functionality instead of their printing characteristics. For example, the softness of the printing plate enables the printing on compressible surfaces such as paperboard and corrugated board, as well as in metallised films or any other type of pressure sensitive coated films and foils. Glass and textiles can also be printed with flexography. A wide variety of inks can also be used, these being either oil-based or water-based. They are typically characterised for having a low viscosity, in the range of 0.05 to 0.5 Pa·s, and a quick drying. They form a ink layer of up to 1 μm (Kipphan, 2001). These characteristics make flexography a widely used technique for printing in the packaging industry.

The potential of flexography printing as a fast printing process for Printed Electronics has been, until now, only demonstrated in a small number of applications. It is been used to print conductive traces (Deganello et al., 2012; Kwak et al., 2010) and transistors (Kaihovirta et al., 2010), and to prepare electrodes in polymer solar cells (Yu et al., 2012). An interesting application was its use to print large-area piezoelectric loudspeakers on paper (Hübler et al., 2012).

2.2.1.3 Offset lithography printing

Offset lithography is currently the most used printing technique, being widely employed to produce large volumes of high quality prints namely newspapers, magazines, brochures, and books. The technique in itself operates on a very simple principle: that oil and water do not mix. The first step of the printing process resides in creating the printing plate. The image to be printed is transferred from the original material to a printing plate, normally using light-sensitive chemicals and photographic techniques, in such a way that the image areas on the printing plate are stimulated to have a strong affinity for oils rather than water, i.e. are oleophilic, and the non-image areas are treated to be oil repelling or oleophobic. During the printing process, the printing plate cylinder is first dampened by water, followed by ink. Whilst the ink adheres to the image area (oleophilic area), the water adheres to the non-image area (oleophobic area). The image is then transferred from the printing plate cylinder to a rubber blanket cylinder and from this one to the printing substrate (Kipphan, 2001; Romano, 1999). Figure 2-8 illustrates the printing principle of offset lithography.

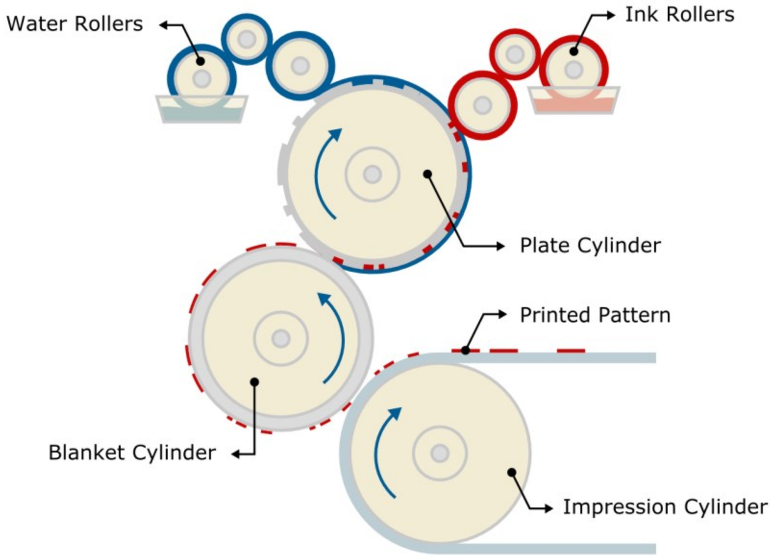

Figure 2-8: Schematic illustration of the lithography offset printing process. Source: Adapter from (Kipphan, 2001).

The term "offset" comes from the fact that the produced print is not directly attached to the target material but first transferred to an intermediary cylinder, the blanket cylinder. The printing plate never actually touches the printing substrate.

The use of an intermediary offset cylinder has the advantage of prolonging the lifespan of the printing plates. When compared to other printing technologies where the printing plate is in direct contact with the printing material, the wear and tear is greatly inferior (Kantola et al., 2009).

The inks used in offset lithography are required to have a high viscosity, paste like behaviour, i.e. a dynamic viscosity in the range of 40 to 100 Pa·s. Also, they must be prepared in a way that the drying components in the ink do not harden while being spread over the ink rollers in the inking unit or at the printing plate and blanket cylinders. Due to the multitude of requirements on the finished printed products and the nature of the substrates, a wide range of inks is available for offset printing. The ink film transferred onto the substrate is extremely thin, having usually a thickness of approximately 0.5 to 1.5 μm (Kipphan, 2001). The biggest disadvantage of offset lithography is related to set-up costs, which are rather high, although the actual printing process is relatively inexpensive.

Standard offset lithography printing processes have already been used to deposit electrically conductive films onto a wide range of flexible materials. Composite structures containing conductive, resistive, dielectric and ferromagnetic layers have also been produced. A brief review is provided in (Evans et al., 2001).

2.2.1.4 Gravure printing

Gravure printing is a mechanically simple process when compared to flexography and offset lithography printing processes, with fewer variables to control. It consists of two cylinders: a gravure cylinder, which carries the image to be printed; and an impression cylinder, which transports the substrate through the printing unit and applies the required pressure to transfer the ink. The gravure cylinder is usually made of steel, and the image elements are engraved in its surface, whilst the non-image areas are at a constant, original level. The engraving processes creates in the gravure cylinder surface a wide number of small engraved cavities (or cells), which contain the ink in order to transfer it to the printing substrate. Each cell can have a different depth, according to the intensity of ink to be transferred. Deeper cells will produce more intensive tones whereas less deep cells will produce less intensive ones. During the printing process, the gravure cylinder is partially immersed in an ink bath and the gravure cylinder cells are continuously flooded in ink. The excess of ink is removed by means of a doctor blade so that the ink remains only inside the cells and the cells walls are free of ink.

A more advanced ink filling method is achieved by using a chambered doctor blade system, which can be advantageous for inks containing highly volatile solvents. The ink is then transferred from the cells to the printing substrate by a high printing pressure and the adhesive forces between the printing substrate and the ink. This step requires a good contact between the printing substrate and the gravure cylinder which can be ensured by using a soft impression cylinder (Kipphan, 2001; Romano, 1999; Søndergaard et al., 2013). The functioning principle of gravure printing is illustrated in Figure 2-9.

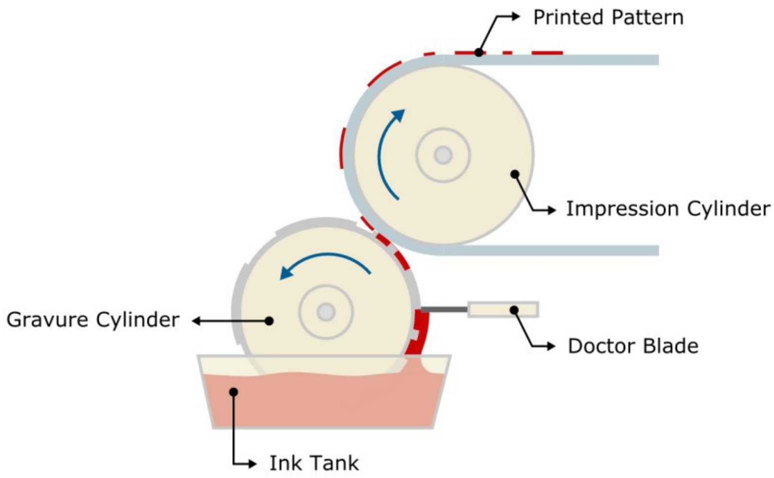

Figure 2-9: Schematic illustration of the gravure printing process. Source: Adapter from (Kipphan, 2001).

Gravure printing is typically used to produce long run printings such as magazines and newspaper inserts, catalogs, postage stamps, plastic laminates, and packaging, since the surface of the gravure cylinder is plated with copper, which is quite expensive (Romano, 1999). The inks used in gravure printing must have a liquid behaviour, with a dynamic viscosity in the range of 0.05 to 0.2 Pa·s, so as to fill the image forming cells of the gravure cylinder at high speeds (up to 15 m/s). From a process point of view, these inks have a simple composition and manufacture process. As a result, the range of workable inks is rather large. Gravure printing also allows a wide range of printing thicknesses, from 50 nm to 5 µm (Kipphan, 2001).

In the context of Printed Electronics, the use of gravure printing for patterning conductive traces has been widely reported (Pudas et al., 2004, 2005; Sung et al., 2010), as well as being demonstrated its applicability, for example, in the production of OLEDs for ligthing applications and displays (Kopola et al., 2009), organic photovoltaic modules (Kopola et al., 2011; Yang et al., 2013), and various sensors (Reddy et al., 2011a, 2011b).

2.2.1.5 Inkjet printing

Inkjet printing is a non-impact printing technology in which droplets of ink are sprayed from a number of nozzles directly onto the printing substrate so as to create an image. The printing system is controlled directly by an image processor in accordance with the specifications of the print job in digital format. Since the process is entirely digitally and electronically controlled, different information content can be printed on every sheet. This has given rise to new and interesting ways of producing printed media, such as print-on-demand, personalisation, and home printing.

Inkjet printing technology has been implemented using numerous designs, each with its own distinctive features. Reviews of the various developments paths can be found, for example, in (Heinzl and Hertz, 1985; Kipphan, 2001; Le, 1998). Notwithstanding the various implementations, inkjet printing can be divided into two basic systems: continuous inkjet and drop-on-demand inkjet. Figure 2-10 provides a schematic diagram of the most relevant inkjet printing technologies.

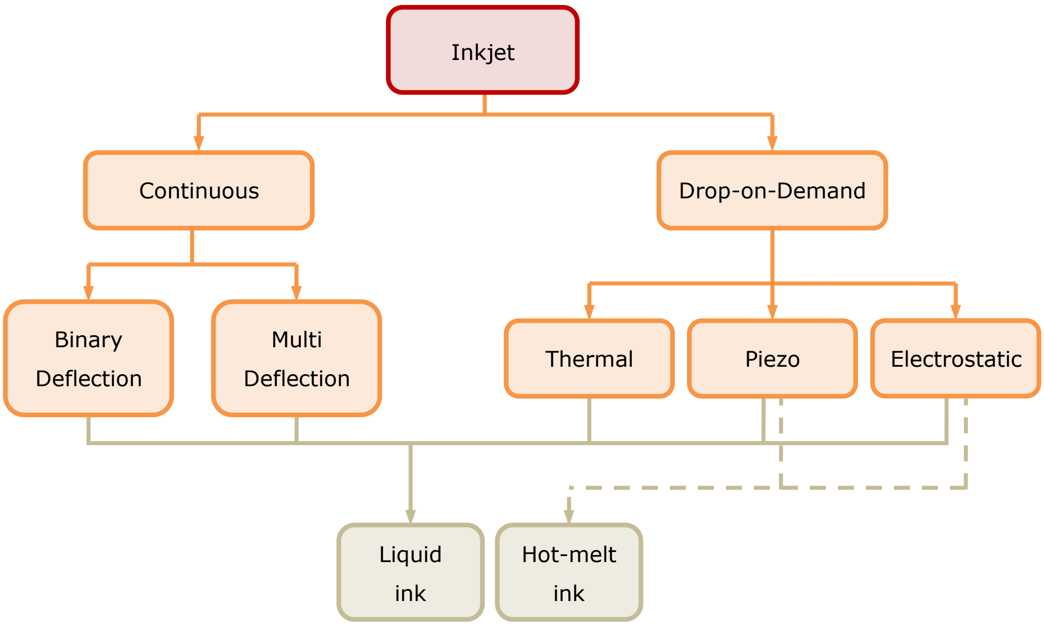

Figure 2-10: Inkjet printing processes. Source: adapted from (Kipphan, 2001).

In continuous inkjet printing, as the name indicates, there is a continuous stream of ink drops during the printing process. These drops are then, in accordance with the image to be printed, either directed to the printing substrate or to a collector for recirculation and re-use. Depending on the drop deflection method, the continuous inkjet system can either be designed as a binary deflection system (Figure 2-11a) or a multiple deflection system (Figure 2-11b). In a binary deflection system, the drops have one of two charge states, i.e. they are either charged or uncharged. The charged drops are directed to the printing substrate, while the uncharged drops are deflected into a gutter for recirculation (or the reverse, when the deflected drops reach the substrate and the undeflected drops enter the recycling system). In contrast, in a multiple deflection system, the drops receive different charges, so that they can be deflected to different directions and transferred to different positions on the substrate. The uncharged drops are, likewise the binary deflection system, deflected to a gutter to be re-circulated.

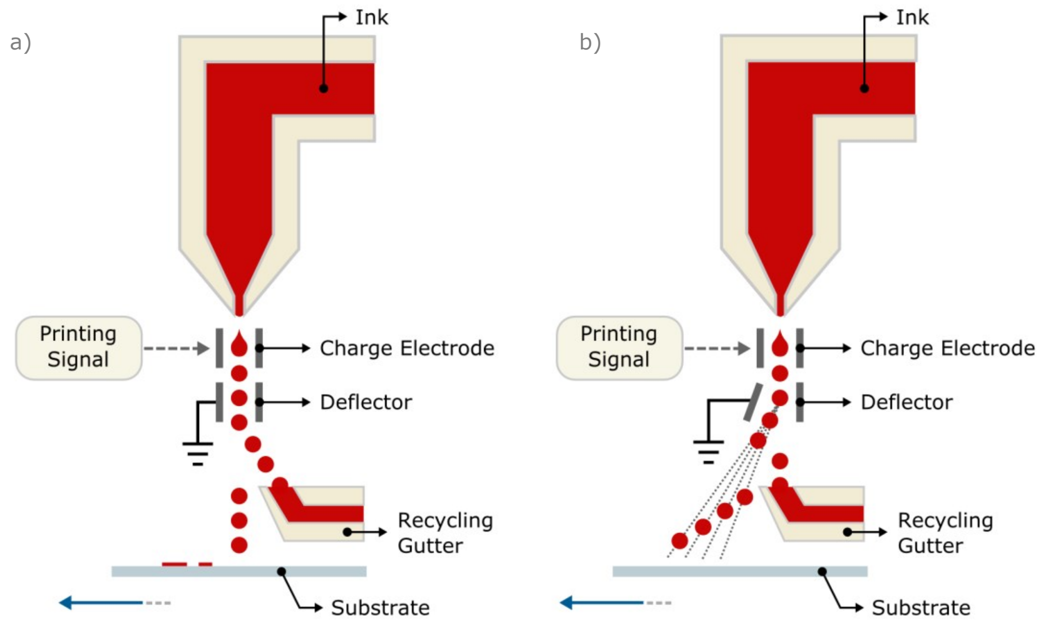

Figure 2-11: Schematic illustration of the functioning principle of continuous inkjet printing: (a) binary deflection system, and (b) multiple deflection system.

In drop-on-demand inkjet printing, the ink drops are only generated when they are required to form the printed image. Hence, there is no need for a deflection or recycling system. Drop-on-demand inkjet systems can be classified according to the process by which the individual ink drops are generated. The processes briefly considered here are thermal inkjet (Figure 2-12a), piezo inkjet (Figure 2-12b), and electrostatic inkjet (Figure 2-12c).

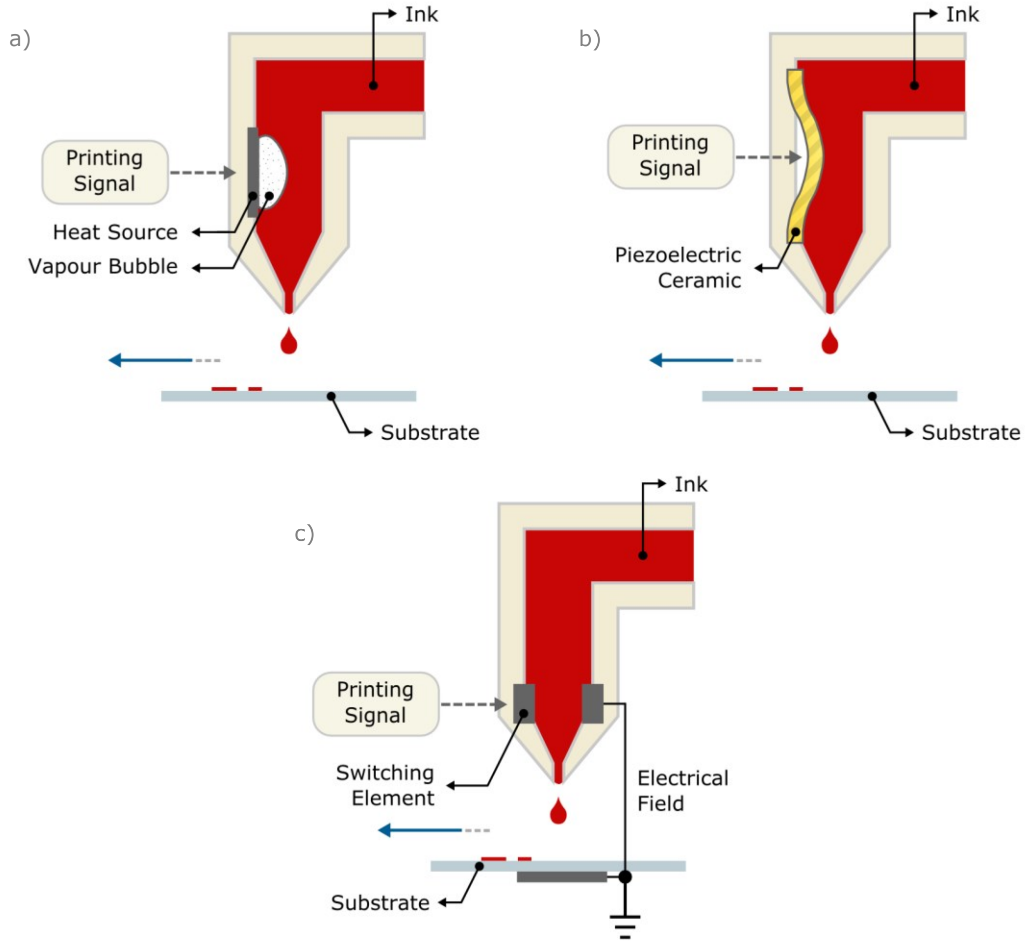

Figure 2-12: Schematic illustration of the functioning principle of drop-on-demand inkjet printing: (a) thermal system, (b) piezo system, and (c) electrostatic system.

The thermal inkjet method is nowadays the most used inkjet printing method on the consumer market. In its essence, the technology consists of an ink chamber with a heater and a nozzle. The ink drops are generated by rapidly heating (few microseconds) the liquid ink inside the ink chamber until it vaporizes, at which point a ink drop is ejected from the nozzle as a result of the pressure exerted by the vapour bubble. The ink chamber is then refilled and the process is ready to start over. In piezo inkjet systems, the ink drops result from a change of volume within the ink chamber due to the deformation of a ceramic piezoelectric material when an electric field is applied. The distortion is used to increase the pressure inside the ink chamber and, as a result, the ink drops are ejected from the nozzle system toward the printing substrate. Electrostatic inkjet in turn, is based on the fact that an electrical field exists between the inkjet system and the surface to be printed, and that by means of image-dependent alterations in the inkjet nozzle system, either the forces can be balanced or the surface tension ratios between the ink and the outlet nozzle can be changed, so that a drop of ink is released as a result of the field forces (Kipphan, 2001; Le, 1998).

One of the most crucial elements in an inkjet printing system, independently of the method considered, is the printing nozzle. Each inkjet system is commonly composed of thousands of high-precision nozzles, typically about 10 micrometers in diameter. The characteristics of the ink drops, i.e. their volume, velocity, and trajectory angle, are directly affected by the nozzle geometry. Even small modifications in the manufacturing process of a nozzle can significantly alter the printing quality. Hence, in order to achieve consistently uniform ink drops, the diameter of each nozzle is fabricated with sub-micrometer accuracy. High resolution printings require small ink drops volume and, as such, the nozzle diameter of print heads has to be relatively small (Hanson, 2009; Le, 1998).

Inkjet technologies typically function with liquid inks with low viscosity (0.001 to 0.03 Pa·s), although piezo and electrostatic inkjet systems can also employ the so called hot-melt inks. If liquid inks are used, the drying process occurs through evaporation and absorption, which can be accelerated through the application of heat. The use of hot-melt ink implies that the drying process is automatically included in the printing process: hot-melt inks must be melted before printing and quickly cool down and solidify upon printing, when in contact with the printing substrate, due to being exposed to ambient temperature. The ink used and its relation with the printing substrate determine the thickness of the ink layer and the quality of the printed image. When liquid inks are used, very thin ink films can be applied, resulting in thickness of approximately 0.5 µm. In the case of being used hot-melt ink, the ink layer thickness ranges between 10 to 15 µm. Both types of ink can comprise dyes or pigments as colorants (Kipphan, 2001; Romano, 1999).

The biggest advantages of inkjet printing, when compared to conventional printing processes, are the possibility to easily change and adjust the printed pattern on a computer without the need to manufacture a physical printing form, and the ability to produce high quality prints, in a variety of substrates, at a relatively low cost. The equipment is also cheaper and more compact, with the added value that multiple print heads can be implemented and used during printing. However, the productivity of these systems is still lower than conventional printing technologies. Inkjet printing is a relatively new technology and presents some limitation with respect to processing speeds and ink formulation.

Noteworthy about inkjet printing is that it allowed the democratisation of printing by enabling desktop printing. Today, inkjet printers are practically within the reach of everyone, being present in most households of the more economically developed countries.

The use of inkjet printing in Printed Electronics is extensive, being reported its applicability in most, if not all, of the applications described previously, from printed memories (Allen et al., 2011) and transistors (Kawase et al., 2003; D Kim et al., 2009), to displays (Chang et al., 1999; Furusawa et al., 2002; Shim et al., 2008; Shimoda, 2003; Shimoda et al., 2003) and photovoltaic cells (Eom et al., 2009; Galagan et al., 2012), including as well RFID modules (Yang and Tentzeris, 2007) and sensors (Lee et al., 2005; Li et al., 2007).

2.2.2 Functional inks

Modern printing inks are commonly formed by four basic components: (1) the colorant, usually a pigment although dyes can also be used, is responsible for conferring the colour to the ink; (2) the binder, whose main purpose is to join the various components of the ink together into a printable film and enable the ink to attach onto the printing substrate; (3) the solvent, used to dissolve the binder and make the ink flow so that it can be transferred to the printing surface; and (4) the additive(s), which is added to manipulate the physical properties of the ink in order to tune it to different situations. In a similar way, functional inks formulations are composed by the same components, with the difference that the colorant element found in printing inks is replaced by a functional element. It is this element that confers the electro-optical properties to the functional ink. The proper formulation of a functional ink involves a complex balance between all these elements. The ink must provide a print film with an adequate cohesion and adhesion to the printing substrate, in addition to possessing the electro-optical characteristics of the functional element. One of the main challenges resides in fact in selecting the adequate additives to improve the printability and processability of the ink without interfering with the electro-optical characteristics of the ink, and thus the key functionality of the printed object. Viscosity, surface tension, and wettability are all critical characteristics in the formulation of conductive inks. The choice of an adequate substrate can also greatly influence the performance of the functional ink.

In general, these inks have the tendency to lose part of their conductivity on porous and uneven materials.

A wide variety of different functional inks formulations have already been developed for Printed Electronics, being commercialised by several companies (e.g. DuPont[3] and SunChemical[4]). Each has their own set of characteristics and uniqueness. They include diverse types of conductive and semiconductive inks, as well as dielectric inks, all of which can be used in numerous applications with various purposes, from conductive traces that mimic the function of electrical wires to constituents of passive and active components, or as dielectric and encapsulating layers. In general, conductive inks are composed of either metallic particles suspended in binders, conjugated conductive polymers, or organic-metallic blends. Silver, copper and carbon, either in the form of pellets, flakes or nanoparticles, are currently the most used elements in the formulation of conductive inks. Semiconducting inks, in turn, are composed of semiconducting organic polymers, inorganic nanoparticles suspended in carrier fluids, or organic-inorganic blends. Dielectric inks are composed of organic polymers, organic polymer thermosets or ceramic-filled organic polymers.

A brief description of the most common types of conductive inks currently available on the market is provided in the following sub-sections. Table 2-3 provides a comparison of the conductivity and resistivity values of the chemical elements commonly used as functional elements in the formulation of these inks.

Table 2-3: Conductivity and resistivity values, at 20 ºC, of functional elements commonly used in the formulation of conductive inks.

	Conductivity (S/m)	Resistivity (Ω·m)
Silver	$6.28 \times 10^{+7}$	1.59×10^{-8}
Copper	$6.01 \times 10^{+7}$	1.66×10^{-8}
Gold	$4.26 \times 10^{+7}$	2.35×10^{-8}
Aluminium	$3.77 \times 10^{+7}$	2.66×10^{-8}
Platinum	$9.44 \times 10^{+6}$	1.06×10^{-7}
Carbon	$2.00 \times 10^{+3}$	3.50×10^{-5}

Source: (EDDY, 2012).

[3] http://www.dupont.com/

[4] http://www.sunchemical.com/

2.2.2.1 Silver-based and nanosilver inks

Silver-based inks are highly conductive and offer excellent flexibility. Indeed, silver is the most conductive metal, with a conductivity of $6.28 \times 10^{+7}$ S/m and a resistance of 1.59×10^{-8} Ω·m at 20 ºC. Beyond the high conductivity, silver also performs better than most metals under oxidative conditions (the oxide layer that is formed during the cure process is conductive), possessing an excellent durability under various conditions. Moreover, silver-based inks present a high mechanical adherence to various substrates, making them ideal for use in a wide range of electronic products, from sensors, flexible displays, photovoltaic cells, and RFID antennas.

Silver-based inks are mostly sold under the form of silver flake inks, though nanoparticles silver-based inks are gaining market share principally due to the fact that flakes tend to be unsuitable for inkjet printing. They are too big for the printer nozzles, frequently clogging them. While flake inks have particle diameters in the range of microns, approximately from 0.5 to 8 µm, nanoparticles inks have nominal particle diameters in the range of 2 to 50 nm (Laakso et al., 2009).

The sintering[5] temperature of nano-sized metal particles is also lower when compared to that of the counterpart metal in the bulk form (Buffat and Borel, 1976; Qi, 2005). Typical sintering temperatures of 100 to 300 °C are required to burn-off the organic additives present in nanoparticles inks and stimulate the sintering process to realise a more densely packed silver layer and form a conductive film of low resistance (Kim and Moon, 2005; Perelaer et al., 2008). The sintering time, naturally, varies according with the composition of the ink used, though, typically, it is only necessary a short period, in the order of 10 minutes. As the printed patterns are heated above the temperature at which the particles lose their organic shell and start showing conductance by direct physical contact, their resistance rapidly decreases. However, conductivities values close to that of bulk silver are usually only reached after sintering the printed patterns at temperatures above 200 to 250 ºC, sometimes as high as 400 ºC (Kamyshny et al., 2011). Obviously, this can end up limiting the choice of substrate. For example, in order for the silver ink to be compatible with plastic substrates the sintering must occur at a temperature lower than 160 ºC (Li et al., 2005). Good film conductivities can, nonetheless, be obtained at temperatures of this order. Dearden *et al.* (2005) reports the formulation of silver-based inks with good conductivity values (2 to 3 times the

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[5] Sintering is a process, typically thermal, for bonding particles together into a coherent, predominantly solid structure via mass transport events that occur fundamentally at atomic level (German, 1996).

theoretical resistivity of bulk silver) at the relatively low temperature of 150 ºC. In turn, Perelaer *et al.* (2008) demonstrated that silver-based inks containing very low amount of organic additives revealed measurable conductivity already after sintering at 80 ºC. Hence, it is of utmost importance to identify an optimum between sintering time and temperature, and the obtained conductivity.

The methods for preparing metal nanoparticles, including silver and gold nanoparticles to be used in conductive inkjet inks, can be divided into two main strategies: (1) top-down and (2) bottom-up. Top-down, or physical methods are generally high-energy methods, in which the bulk metal or the microscopic particles are converted to nano-sized particles. These methods are expensive, energy-consuming, and require sophisticated equipment. In bottom-up methods, in contrast, nanoparticles are built up from metal atoms and nuclei, which are formed either from precursor ions and molecules with the use of a proper reducing agent, or from precursor molecules by their decomposition. Bottom-up preparation of metal nanoparticles is usually performed in a liquid medium, which can vary from water to polar and non-polar organic solvents and ionic liquids (Kamyshny et al., 2011).

The biggest drawback of using silver-based inks is undoubtedly its cost. Silver has a high price, making it expensive for many applications. Also, its price is very susceptible to rate fluctuations, making it hard to estimate production costs. Nonetheless, for now, silver is the most common precious metal used in the development of conductive inks for Printed Electronics, existing a wide range of inks formulations compatible with the printing processes addressed in this chapter. Its high performance makes it a cost effective material that it is not easily replaced.

2.2.2.2 Copper-based and nanocopper inks

Copper-based inks present a conductivity roughly compared to that of silver-based inks but at a more affordable price. Furthermore, likewise silver-base inks, copper-based inks have a strong adhesion to a wide variety of substrate materials, making them also suitable for printing electrical circuits in a broad range of electronic products. The drawback is that copper is easily oxidised and, unlike the silver oxide, the copper oxide is insulating. As a result, copper becomes less conductive as it oxidises. Hence, protective agents capable to retard the oxidation process are of utterly importance in the formulation of these inks. Moreover, it is advisable that printed copper patterns be protected by an overcoat in order to prevent the long term oxidation of the copper metal to copper oxide.

Printed copper patterns exhibited metal-like appearance and, as they dry become more conductive. Sintering greatly improves the conductivity of these as it speeds up the drying process and prevents the initial development of the copper oxide layer that is typically formed during the drying process. High conductivity values close to that of the bulk material require sintering at temperatures in the order of 350 ºC (Haque et al., 2013; Park et al., 2007). However, good sintered copper structures can be achieved at temperatures of 250 ºC (Lee et al., 2009). Akin to silver-based inks, the use of temperatures lower than 160 °C allow the application of copper-based inks to plastic substrates. Naturally, a compromise must be achieved in terms of conductivity.

2.2.2.3 Gold-based and nanogold inks

Gold possesses a high conductivity ($4.26 \times 10^{+7}$ S/m at 20 ºC), only slightly inferior to that of silver and copper, coupled with the added value of having a high stability. Indeed, gold is one of the most non-reactive metals, only surpassed by platinum, being unaltered by factors like heat and humidity. It has a general resistance to oxidation and corrosion, never reacting with oxygen. Hence, it never degrades, rusts or tarnishes over time. It is these characteristics, together with gold's unparalleled ductility and malleability that make it an essential element in the field of electronics. It has a high work function and serves as the single most reliable metal in the field.

The single and biggest drawback of gold is its high cost. If silver prices are already considered high, with various efforts underway to identify more cost effective alternatives such as copper-based inks, gold cost are even higher and equally prone to market fluctuations. Nonetheless, there are several market opportunities for gold-based inks, where the benefits of using gold can potentially outweigh the higher cost. These include, for instance, the development of sensors for environmental and medical applications (Bonfil et al., 2000; Laschi et al., 2006).

2.2.2.4 Aluminium-based inks

Aluminium has a conductivity of $3.77x10^{+7}$ S/m, being alongside with silver, copper, and gold as one of the best conductors known to date. It is the most abundant metal on Earth and has an excellent resistance to corrosion. However, this resistance results from a thin surface layer of aluminium oxide that is rapidly formed when the metal is exposed to oxygen, which creates a physical barrier to corrosion and further oxidation. The problem is that the aluminium oxide layer is non-conductive, and thus greatly tampers the conductivity of aluminium-based inks. Hence, like copper-based inks, aluminium-based inks require an overcoat after application. On the positive side, aluminium is cheaper than copper.

Aluminium has been widely employed, under the form of thin foils, in the manufacture of RFID antennas of so-called traditional RFID tags through conventional printed circuit subtractive processes. However, under the context of Printed Electronics, aluminium-based inks seem to be deprived in favour of silver and copper-based inks. The low cost of aluminium-based inks appears to not justify its use considering the performance lost. A report from the VTT Technical Research Centre of Finland details the development of a low work function aluminium-based ink, along with its advantages, that intends to be a substitute for printable silver inks and pastes in a wide range of printed electronics applications. According to the authors, it is ideal for organic photovoltaic and OLED display components, organic field-effect transistors (OFET), RFID antennas, and sensor applications (Rupprecht et al., 2012).

2.2.2.5 Platinum-based inks

Like gold and silver, platinum is a noble metal. It has a conductivity of $9.44x10^{+6}$ S/m and it is the least reactive metal on the periodic table of elements. Hence, platinum exhibits an excellent resistance to oxidation, not being oxidised at any temperature, and to corrosion, being resistant to acids. However, as one of the rarest elements on earth, its price is also high, approximately two orders of magnitude higher that the cost of silver. Hence, platinum-based conductive inks are expensive and their cost makes then prohibitive for certain, if not most applications. Platinum-based inks should only be used if silver-based inks have been tried and found not to be appropriate or when known not to work, for example, due to the anodic oxidation of silver.

The use of platinum-based inks has been mainly reported in the fabrication screen printed biosensors for analytic determinations in food and environmental analysis. Platinum is considered to be a very suitable material for the detection of hydrogen peroxide (H_2O_2), and various biosensors are based on oxidases as biological components, i.e. on enzymes that catalyse oxidation-reduction reactions involving molecular oxygen (O_2). The measurements are them based on the produced H_2O_2 that results from the enzymatic reaction at the platinum electrode (Chemnitius and Bilitewski, 1996; de Mattos et al., 2003; Erlenkötter et al., 2000; Fernández Romero et al., 1998).

2.2.2.6 Carbon-based inks

Carbon-based inks, although conductive, have high resistivity values. Hence, they tend not to be sufficiently conductive for most Printed Electronics applications, not being suitable, for instance, for patterning electrical circuits. Moreover, carbon-based inks have a propensity to suffer from poor adhesion to substrates and poor cohesion within the ink film, resulting in poor flexibility and rub resistance, which can end up limiting its use in applications that require significant handling. Nonetheless, carbon-based inks have their applicability in the field of Printed Electronics. They allow the creation of resistors and, when used in conjunction with other conductive inks, namely silver-based inks, enable the fabrication of conductor/resistor circuits that can be printed on numerous low temperature substrates. Their low cost also makes them ideal for printing highly sensitive biosensors suitable for the cost competitive market of disposable sensors (Boujtita et al., 2000; Crouch et al., 2005; Hart et al., 2005; Wang et al., 1996).

Carbon-based inks conductivity can be increased if applied in multiple layers and they typically only take 10 to 15 minutes to dry at ambient temperature, and thus to achieve maximum conductivity.

2.2.2.6.1 Carbon nanotubes

Carbon nanotubes (CNTs) are tubular shape materials, with a diameter measuring on the nanometer scale, consisting of carbon atoms arranged in a hexagonal array. Since their discovery in 1991 by Iijima (1991), carbon nanotubes have attracted a great deal of attention as a highly conductive form of carbon, being the focus of considerable research in various fields, from electronics to energy and biological applications (Endo et al., 2008).

Carbon nanotubes can be produced with several structures, differing in length, thickness, type of helicity (armchair, zigzag or chiral), and number of layers (single-walled versus multi-walled) (Saifuddin et al., 2013; Thostenson et al., 2001). Despite being all formed from essentially the same graphite sheet, the electrical characteristics of each structure differ depending on these variations, either acting as a metal or as a semiconductor (Wildoer et al., 1998). Overall, carbon nanotubes demonstrate a unique combination of strength and stiffness of carbon fibres with the specific thermal and electrical conductivity of metals. They are one of the strongest materials discovered to date, have a good chemical and environmental stability, and a high thermal conductivity. The electronic properties are also extraordinary, presenting a high electrical conductivity, comparable to copper.

The main drawbacks that hinder the use of carbon nanotubes are essentially related to their synthesis, namely the limited understanding of it, as well as of their properties. This leads to poor processability, difficult structure control, and poor dispersion stability (Saifuddin et al., 2013; Vigolo and Hérold, 2011). Moreover, as carbon nanotubes produced using the currently available methods result in a mixture of conducting and semi-conducting forms, their overall conductivity levels also tend to be unreliable. As a result, broader applications of carbon nanotubes to real world problems have gone largely unfulfilled, including in the field of Printed Electronics. Nonetheless, reports of carbon nanotubes with high conductivities comparable to metals are available (Ryu et al., 2012), as well as of various inkjet conductive inks based on carbon nanotubes (Beecher et al., 2007; Fan et al., 2005; Kordás et al., 2006; Wei et al., 2007).

2.2.2.6.2 <u>Graphene</u>

Graphene structure consists of a single layer of carbon atoms, with one-atom thick, arranged in a regular hexagonal pattern. Roughly, it can be thought of as an unrolled carbon nanotube. The first theoretical studies on graphene date as long as 1947 (Wallace, 1947), although the name "graphene" was only first mentioned in 1987 by Mouras and co-workers (Mouras et al., 1987) to describe the graphite layers that had various compounds inserted between them. For many years, and despite being known as an integral part of 3D carbon-based materials, graphene was considered a purely academic material, presumed not to exist in the free-standing form. When in 2004, Andre Geim and Konstantin Novoselov, from The University of Manchester, manage to isolate the first free-standing graphene sheets using mechanical exfoliation (Novoselov et al., 2004; Novoselov, Jiang, et al., 2005), followed by the initial characterisation tests (Novoselov, Geim, et al., 2005; Zhang et al., 2005), graphene emerged as a much desirable material. The unique electronic and mechanical properties of graphene attracted the interest from the scientific and industry communities, resulting in an increase of research in the field (Novoselov et al., 2012).

Graphene is the thinnest and strongest material known to date. It is also the most conductive form of carbon, with high electron mobility at room temperature. Moreover, it has a high optical transparency and, when oxidised does not form an insulating oxide film. These unique properties, combined with graphene ease of preparation, make it suitable for a broad number of applications in various technological fields, from supercapacitors and field-effect transistors, to sensors and transparent conducting films (Geim and Novoselov, 2007; Geim, 2009; Grande et al., 2012; Novoselov et al., 2012; Zhu et al., 2010). Graphene represents a potential breakthrough in the field of Printed Electronics by enabling the production of conductive inks with a high conductivity and flexibility at a cost far below of competing silver-based inks. Graphene-based inks can be tailored to function on a wide variety of printing technologies and substrates. Moreover, they can be made to present a vastly improved handling characteristic relative to carbon inks, being extremely bendable when printed, and suffering only a minimal drop of conductivity after a multitude of folds. Various graphene-based inkjet inks have already been demonstrated (Huang et al., 2011; Torrisi et al., 2012).

2.2.2.7 Organic inks

Conjugated conducting polymers are a class of materials with a unique set of properties (see for instance, (Chandrasekhar, 1999; Chilton and Goosey, 1995; Inzelt, 2012)). They possess the electronic properties of metals and semiconductors combined with the mechanical characteristics of polymers. They are lighter, more flexible, and less expensive than inorganic conductors. Moreover, their properties can be selectively fine tuned through the introduction of appropriate molecular dopants. According with the synthetic method used to prepare these materials, conjugated conducting polymers can either be classified in chemically polymerised materials or electrochemical polymerised (Sadki et al., 2000; Toshima and Hara, 1995).

However, most conducting polymers are insoluble, which greatly hinders their processability. In the cases where conducting polymers can be made soluble, usually by functionalising the polymer backbone, they are excellent candidates for the formulation of electro-optical functional inks for Printed Electronics applications. For example, conductive polymers such as polyanilines (PANi), polypyrroles (PPy) and polythiophenes, in particular poly(3,4-ethyl-enedioxythiophene) (PEDOT), have been widely investigated and optimised for being printed with various printing methods, namely the ones addressed in this chapter (Knobloch et al., 2004; Srichan et al., 2009; Weng et al., 2010). Another challenge in the development of appropriate conducting polymers-based inks is related with the ability to produce materials of sufficient conductivity. In general, conjugated conducting polymers-based inks tend to have a conductivity value lower than metal-based inks, making them not the most ideal for certain electronic applications. Annex A addresses in more detail the characteristics and performance of conjugated conducting polymers, focusing also on the optoelectronic properties of these materials.

Current applications of conjugated conductive polymers include, for example, organic light-emitting devices (Dai et al., 2001; Friend et al., 1999), organic solar cells (Brabec et al., 2001; Eom et al., 2009), organic transistors (Chason et al., 2005), electrochromic displays (Andersson et al., 2007; Argun et al., 2004), and biosensors (Gerard et al., 2002).

2.2.3 Considerations on the environmental impacts of Printed Electronics

With the panorama that in a near future the number of Printed Electronics devices might be very well high, concerns over their environmental impacts merits consideration. Electronic waste is today a serious and complex environmental problem that undermines the ecological and economic sustainability of communities worldwide (Grossman, 2007; Schluep et al., 2009). The rapid technological advances witnessed in the last two decades brought faster and smaller electronic devices. However, it also has led to devices that easily become obsolete. The rapid changes in technology, along with recurrent changes in media formats, planned obsolescence and constant falling prices with each new development, resulted in shorter product lifetimes and consequently in a fast-growing stream of electronic waste around the world. Waste that results not only of the disposal of these products at the end of their remarkably short lives but as well as from the mining processes of the raw materials that go into these devices and from their manufacturing processes (Grossman, 2007).

Electronic devices contain a wide variety of materials in their components, many of which are toxic and hazardous. For example, tin, lead, mercury and cadmium are common elements present in all electronic devices. While some valuable metals such as gold and silver are separated from the products, mainly through manual disassembly in poorer communities of China and India (Schluep et al., 2009), a surprisingly large amount of the waste ends up in nature, causing environmental degradation and health problems. Hence, the effective recycling of these metals and materials is crucial to prevent such situation, as well as to keep them available for the manufacture of new devices.

With the mass production of Printed Electronics, an increasingly higher number of digital devices will be produced irremediably resulting in an increase in the amount of future waste, even more if taken into account that some devices are being targeted as disposable due to their low cost. On the positive side, the fact that printing is an additive process is an advantage as fewer raw materials will be wasted. Keskinen and Valkama (2009) argue that early identification of the environmental and health impacts associated with Printed Electronics is advantageous in that proactive measures can then be taken to diminish them in each step of the product life cycle as well as to ensure the success and future of Printed Electronics. However, there is a lack of comprehensive studies and thus a lack of knowledge to the extent in which printed electronics applications can be reused and recycled, and what possible environmental and health risks they could pose if ended up in a landfill. These concerns are in part related to the fact that Printed Electronics comes from the use of novel materials (e.g. nanoparticles).

Power consumption is also an important issue. Despite the noted low consumption of Printed Electronic devices, as they become more prevalent, the overall need for power supply will inevitably increase. The development of complementary printed energy harvesting technologies is hence a necessity.

2.3 Personal Fabrication and the democratisation of technology

Personal Fabrication refers to the ability of ordinary people to design and produce their own products using digital fabrication tools directly from their homes. By making accessible the capabilities of manufacture machines tools into the home, it enables users, even those without any special skills or training, to create three-dimensional (3D) physical structures, as well as electronic circuits, sensors, and actuators that can be incorporated into these structures, thus creating complete functioning digital systems, from digital designs. Indeed, digital fabrication tools enable individuals to manipulate atoms as easily as they manipulate bits. It brings the programmability of the digital worlds we invented to the physical world we inhabit. To Gershenfeld (2005), a chief advocate of the value that Personal Fabrication can have in the democratisation of technology and innovation, the goal is to give back to users the control of the creation of technology, while fulfilling their individual desires. It provides the means for almost anyone to make almost anything. Instead of being limited by what is available in stores and being obliged to purchase something that someone else believed they wanted, individuals become limited only by their imagination. Moreover, being technology developed by and for them, it undoubtedly reflects better their needs and wishes. They can develop exactly what they want. The enjoyment of the innovation process is another important aspect. For certain individuals, the creation and learning process is of extreme value. Nonetheless, individuals do not have to develop everything on their own. They can benefit from innovations developed and freely shared by others (Anderson, 2012; Hippel, 2005). Overall, Personal Fabrication is an empowering movement, enabling individuals to personally program the construction of their physical world as they see fit. Hence, it aims at democratising not just the use of technology but its development as well.

To a certain extent, the vision of Personal Fabrication is today already a reality. Although most people do not have (yet) at their homes the required machine tools to make their own products, they indeed can have access to them, whether through one of the thousand makerspaces and hackerspaces that exist throughout the world, or through a Fab Lab (see Box 2-3).

Box 2-3: Makerspaces, Hackerspaces and Fab Labs.

Makerspaces and Hackerspaces

Makerspaces (Makerspace, n.d.) and hackerspaces (Hackerspaces, 2012) are community-based and community-managed physical places where people can gather and share their experience and expertise, as well as work on their projects. Both seek to promote active participation, knowledge sharing, and collaboration among individuals, through open exploration and creative use of technology. As such, they are spaces for experimentation, testing, and development of new objects and concepts. More than providing the hardware tools and manufacturing equipment, they provide the learning environment and the necessary support for individuals to develop their projects based on their own interests. Each space is unique in the sense that it emerge directly out of a local community, and it is maintained by that specific community, from loosely-organised individuals sharing a space and tools, to profit companies and organisations affiliated with or hosted within schools or universities. Hence, commonly each space has its own ideology and organisational model. They are all completely independent from each others, though collaboration between spaces is quite common.

Fab Labs (Fabrication Laboratories)

Fab Labs (Center for Bits and Atoms, n.d.), as the name indicates, can be regarded primarily as fabrication laboratories, providing communities, businesses, and entrepreneurs the necessary manufacturing machines and tools to turn their ideas and concepts into reality. The concept was developed by Neil Gershenfeld (see (Gershenfeld, 2005)), from the Center for Bits and Atoms (CBA) of the Massachussets Institute of Technology (MIT), initially with the aim to explore the implications and applications of personal fabrication in those parts of the world that cannot easily have access to tools for fabrication and instrumentation. The first Fab Labs were created in 2002, in rural India, Costa Rica, northern Norway, inner-city Boston and Ghana. A distinctive feature of Fab Labs is that they all share at their core the same hardware and software capabilities, making it possible for people and projects to be easily disseminated across them. A brief description of the core manufacture machine tools that compose a Fab Lab is provided in Box 2-4. Noteworthy, that this is a minimal set of necessary tools, and by any means the structure of Fab Labs is static and rigid. Fab Labs are as well based on a strong feeling of community, being supported by a global Fab Lab association (International Fab Lab Association, n.d.). From the dissemination of the Fab Lab concept to being the connection point between the various Fab Labs across the world, the Fab Lab association objectives also comprise the promotion of collaboration among labs, the share of expertise, the brainstorm of ideas, and the spread of research. To a certain extent, Fab Labs can be seen as an organisational evolution of the hackerspace structure, focusing on the manufacture of custom built objects.

It is expected that with the continuous evolution of technology, personal digital fabrication technologies will likewise mature, becoming more functional, reliable and, more importantly, easy to use and affordable. As a result, they will become progressively more common in businesses, schools and consumers homes, ultimately tipping Personal Fabrication from a movement of pioneers and early adopters to mainstream, as an everyday activity done by everyone. It is at that point that the unique benefits of Personal Fabrication will become truly evident. For now, the adopters of Personal Fabrication are mainly technologically sophisticated hobbyists, commonly called makers (see for instance, (Anderson, 2012)), which are more interested in the technology itself and its capabilities, that in its design and ease of use. They are the ones pushing Personal Fabrication forward. However, this does not mean that the first effects of Personal Fabrications are not already noticeable. Digital fabrication technologies are already giving a great number of makers the capability to produce their own personal objects.

The internet is also playing an important, not to say fundamental, role in the Personal Fabrication movement. Makers are not only making what they think is relevant for them but also making it accessible to others. Online public repositories are being used to share worldwide the digital blueprints of physical objects being created by its makers. The open licenses under which most of the blueprints are made available online allow other makers not only to freely download and fabricate the objects but also to modify them and create derivatives as they please. The most well-known example of such repositories is the Thingiverse[6].

Interesting as well is the fact that digital fabrication technologies are enabling makers to transform the objects they create into products and goods outside the traditional manufacturing model. Again, the internet has a crucial role by allowing

[6] http://www.thingiverse.com/

them to reach potential consumers, and through websites such as Kickstarter[7] and Indiegogo[8] it becomes possible, by means of crowdfunding, to secure the necessary resources to move from the prototype stage to production. Consequently, we are witnessing an increasingly bottom-up entrepreneurship, associated with the emergence of numerous lightweight factories, as well as the expansion of micro production and mass customisation (Anderson, 2012; Mota, 2011). As Anderson (2012: 50) points out, "*manufacturing new products is no longer the domain of the few, but the opportunity of the many*".

Box 2-4: Fab Labs core manufacture machines technologies. Source: Based on (Gershenfeld, 2005).

Laser Cutter

Laser cutting is a subtractive process that uses a high intensity focused beam of light to cut out shapes in a wide variety of material according to the digital information provided. Desktop laser cutters can cut almost all non-metallic materials, although they are not safe to use with materials that emit dangerous fumes when burned, such as certain plastic materials. The most common kind of desktop lasers cutters work with a carbon dioxide (CO_2) laser, i.e. they uses carbon dioxide as the amplifying medium. As the cutting tool is a beam of light, it can move very quickly, providing fast cutting speeds as well as being capable of narrow cuts, enabling amazing levels of detail and precision. Laser cutting can be so accurate that the cut shapes can be made to snap together, allowing the quick assembly of complex 3D structures. At low power, laser cutters can be used to mark, through engraving, the processed material.

Water Jet Cutters

Water jet cutters work in a similar way to laser cutters. In this case, a highly focused and pressurised stream of water containing tiny abrasive particles is used as the cutting tool. It is these particles that are responsible for the cutting. When they are accelerated to the speed of the jet, they gain so much energy that they become capable of cutting through just about anything. As a result, water jet cutters are capable to cut materials that laser cutters cannot, namely hard

[7] http://www.kickstarter.com/

[8] http://www.indiegogo.com/

materials such as metals and stone with several centimetres thick. The nature of the cutting stream also makes it capable of making fast and fine cuts with tight tolerances for complex shapes. Water jet cutting is also a preferred solution when the materials being cut are sensitive to the high temperatures generated by other cutting methods.

Sign Cutters

Sign cutters, also known as vinyl cutters, use a computer controlled sharp blade to perform precise custom shape cuts out of thin sheets of materials like paper, cardstock, and vinyl. It is also possible to use then to cut thin copper sheets in order to quickly make functional flexible circuits. The applicability of sign cutters is, hence, limited to the materials that the blade can cut through. Sign cutters are relatively cheap and are widely available at craft stores.

Computer Numerical Control (CNC) Milling Machines

In CNC milling, a high speed rotating cutting tool called an end mill, similar to a drill bit, is used to mill, cut and carve precise designs into a broad range of large dimensions materials. Unlike laser cutters and water jet cutters, CNC milling machines can precisely contour and cut three-dimensional shapes. Normally, the cutting tool can move in its three axes, i.e. left-right, front-back, and up-down. In more advanced milling machines, the milling head as well as the material being cut can also be rotated, resulting in four, five and even six-axis milling machines. Naturally, this provides extra flexibility during the cutting process, enabling more complex cuts. The position of the tool is driven by motors that provide highly accurate movements.

There is a wide variety of end mills, each appropriated for a specific type of cut or material. Multiple passes using different end mills allow highly complex curves to be perfectly carved out of different materials from foam to wood to steel. CNC milling machines revolutionised the machining processes by allowing the rapid realisation of complex cuts with extremely high accuracy, something that otherwise could not be easily duplicated by hand.

CNC milling machines exist in various sizes, being the biggest as big as a warehouse. Personal CNC milling machines, as expected, are characterised by equipment whose size, capabilities, and price make it useful and affordable for individuals. Moreover, they are made to be easily operated by end-users without professional training in CNC technology. CNC milling machines, even small ones, are in particular ideal for creating large batches of items.

Printed Circuit Board (PCB) Milling Machine

PCB milling machines are high precision (micron resolution), two-dimensional, desktop size milling machines used to create circuit traces in pre-clad copper boards by removing the undesired areas of copper. PCB milling is a non-chemical process, in contrast to the etching process commonly used in the creation of PCBs, and as such it can be completed in a typical office or lab environment without exposure to hazardous chemicals. However, in mass production, PCB milling is unlikely to replace etching, being currently regarded essentially as a rapid PCB prototyping process.

Three-Dimensional (3D) Printers

3D Printing is an additive manufacturing process that allows the creation of three-dimensional objects with the most diverse shapes and sizes from a digital model. There are several 3D printing processes that can be implemented to print an object. The most commonly used in commercial 3D printers follow one of the subsequent basic approaches: (1) One approach, called selective laser sintering, involves the use of a laser to selectively harden layers of liquid or powder resin in a bath (or bed). The laser sequentially plots cross-sectional slices of the model as the emerging object is lowered into the bath of raw material, until completed. An advantage of this process is that the raw material also serves as support structure for partially completed objects, thus allowing the construction of highly complex objects. (2) A second approach, to a certain extent similar to the first one, uses a liquid binding material to fuse a powder resin in a bath. An inkjet print head is used to deposit the liquid binder onto the fine powder, selectively fusing the powder where the printed droplets land. Hence, the object is created one layer at a time by repetitively spreading and fusing layers of powder. This technology allows the printing of full colour objects by using equivalent coloured binder liquids and, as in the previous approach, the unfused powder serves as well as support structure for partially completed objects. (3) The last approach addressed, called fused deposition modelling, consists of extruding a thermoplastic material from a movable print nozzle, by melting it, into a chamber that is slightly cooler that the melting temperature of the thermoplastic. As the thermoplastic material is extruded, it hardens almost immediately, forming the various layers that compose the final object. Personal 3D printers typically employ this approach mainly due to its simplicity and easy implementation. The biggest disadvantage of this process is that during the printing process there are no support structures for the object being printed and, as such, the object must sustain itself. Also, it is not possible to create objects composed by various independent parts or with moving parts, at least already assembled.

3D printing is mainly used for prototyping and distributed manufacturing since its slow printing speeds make it not feasible for mass-manufacture. Hence, 3D printing can be regarded essentially as a complementing process to traditional subtractive manufacture methods rather than trying to replacing them. It must also be pointed that 3D printing is not limited to plastic materials, as there are already printers capable to print other materials such as glass, steel, bronze, gold, and even cake frosting.

From the digital manufacture technologies described in Box 2-1, 3D printing is the one that raises the most interest due to its possible applications and, in the specific context of this thesis, its possible association with Printed Electronics. One of the limitations of 3D printing, at least for now, is that it can only make unanimated objects. If the object is, for instance, to have movement or be able to show digital information, active components such as motors and displays screens, along with the required microcontrollers and necessary wiring, have to be assembled after the object is completed. Ideally, the integration of these components would be done at the same time as the object is being printed. In the same way that common inkjet printers have several ink cartridges for different colours, 3D printers would have multiple cartridges for different types of materials and functional inks. This would not only enable the printing on-the-fly of objects composed by various materials, from plastic, metal and wood pulp to even food (Periard et al., 2007) or biological tissue (Mannoor et al., 2013), but also would made possible for electronic circuits and components to be directly printed into the mechanical structure of the objects being created (see (Willis et al., 2012)). In the long term, this would allow individuals to fabricate (print) their own "invisible" digital devices and assemble their own "Internet of Things", from the comfort of their homes.

The integration of functional inks into a 3D printer is, ultimately, the route towards making a programmable personal fabricator that will be able to make anything, including itself. It will be a self-reproducing machine (Gershenfeld, 1999, 2005).

2.4 Concluding remarks

Weiser's vision of Ubiquitous Computing is unquestionably a compelling vision of the future. It inspired numerous scholars, becoming a research endeavour embraced by many areas of computer science. It entailed a new paradigm of interaction between humans and computers. The user is placed at the centre of a new way of understanding and designing computer systems. Computer-embedded devices become a natural interface between humans and their environment and, as so, human-computer interaction is seen as a by-product of natural human activities. Technology would assume a liberating role, inspiring calmness.

The technological advances witnessed since Mark Weiser first introduced the concept of Ubiquitous Computing already originated novel paradigms of computation, supported by constant access to information and computational capabilities. It has also changed the way people interact and use the computer, at the same time that created a culture that is substantially more receptive to the deployment of digital technologies. However, today's digital devices are still very present and visible. Hence, it is of paramount importance the exploration of materials and fabrication technologies capable of merging seamlessly the digital information with the physical objects.

Printed Electronics promises to revolutionise the existing electronics field by enabling the mass production of low-cost, lightweight, flexible digital devices. It represents a ground-breaking new type of electronics that opens up entirely new markets for applications with novel form factors. The intention is not to replace the traditional silicon-based electronics (although in certain applications this might end up happening) but to create disruptive market opportunities that until now were not practical with traditional silicon-based electronics. Indeed, Printed Electronics enables a new set of opportunities and possibilities for products by allowing the incorporation of electronic functionalities into artefacts where it was previously unavailable, transforming lifeless objects into sensing, interacting interfaces capable of reacting and exchanging information with users and the environment.

Conventional printing technologies are already being successfully employed in the fabrication of low-cost Printed Electronics devices. The most commonly used technologies are: screen printing, flexography printing, offset lithography printing, gravure printing, and inkjet printing. Each has its own strengths and limitations in regard to the production of Printed Electronics.

When compared to subtractive fabrication methods of traditional silicon-based electronics, printing-based fabrication techniques enable high throughputs, are cheaper and simpler to implement, and allow the use of various types of substrates. However, they are also highly dependent on the ink formulations. The inks used must provide a print film with an adequate cohesion and adhesion to the printing substrate whilst maintaining the electro-optical properties of the functional elements. Indeed, formulation of adequate and cost-effective functional inks is one of the main limiting factors to the widespread of Printed Electronics. Others being the devices low performance and lifetime, with products often falling short on the requirements needed for commercial use.

Silver-based inks, whether in the form of flakes or nanoparticles, are usually the preferred option, essentially due to their best performance, for printing Printed Electronics devices. While the high cost of silver-based conductive inks often makes them a solution too expensive for many applications, lower cost alternatives are in general either insufficiently conductive (e.g. organic inks), or lack in flexibility and handling characteristics (e.g. traditional carbon inks). Naturally, there is a constant research for new inks formulations, with graphene currently being praised as a material capable to revolutionise the field of Printed Electronics by enabling the fabrication of high-performance, low-cost conductive inks suitable for most applications.

From the printing technologies described, inkjet printing is one of the most attractive and versatile technologies for the fabrication of Printed Electronics devices. It enables a wide range of components to be printed on the moment, given that the fabrication process is all digitally controlled, from transistors and sensors to photovoltaic cells and displays. Moreover, it can be used both for prototyping and mass production. However, the productivity of these systems is lower than conventional printing technologies.

The combination of digital fabrication technologies, in particular of 3D printing, with inkjet printing of conductive inks offers an interesting new approach to the design and making of objects, unleashing new fabrication methods and product ideas. In a similar way as inkjet printers have several cartridges for different colours, 3D printers will have multiple cartridges for different materials, enabling not only the printing of objects with multiple colour combinations but more interestingly, the printing on-the-fly of various types of input materials and functional inks. Electronic circuits and components will be directly printed into the mechanical structure of the objects being created thus becoming an integrating part of these and not just an assembled element. Both structural and functional elements will be printed as one. This ultimately will be the route towards making anything, including a self-reproducing machine (Gershenfeld, 1999, 2005).

Interestingly enough, not only professionals will have access to these sophisticated manufacturing technologies. It is foreseen that the evolution of 3D printers will follow a similar path as seen with computers and inkjet printers. In an analogous way to the eras of computing described in section 2.1, it can be said that 3D printing is now entering the second era. It is starting to become more personal and affordable to the public. Professional 3D printers are still expensive, mainly accessible to the public through online fabrication services although cheaper solutions aimed at home use are emerging. These, for now, are still characterised for being rudimentary and hard to use. Given time, 3D printers will become cheaper, easy to use, and more reliable. They will appeal to the consumers that have no special training, and soon after, they will become equally ever-present as today’s personal computers and printers. As seen with personal printing and mass printing, it is expected that Personal Fabrication technologies will not replace mass manufacturing, especially for large, complex products or for commodity products. Instead they will supplement it, allowing users to fabricate custom made products that reflect their specific needs.

One of the notorious achievements of inkjet printing was the democratisation of printing. The affordability of desktop inkjet printers made it possible for ordinary people to print whatever they wanted from the comfort of their homes. With the materialisation of Personal Fabrication, it is the democratisation of innovation and technology that is being embraced. The possibility of ordinary people developing their own embedded digital devices and making their own vision of the Internet of Things come to life is as well a fascinating one. It can lead to interesting and unpredictable developments and ultimately shape the reality of Ubiquitous Computing depending on their needs.

3 Printed Electronics Displays for Novel Visual Information Solutions

3.1 Images as information

A key focus of this thesis is the development of digital devices capable of presenting dynamic visual information. As we attempt to move closer to Weiser's vision of Ubiquitous Computing and the notion of Calm Technology, it becomes increasingly important to develop and implement technologies capable of displaying dynamic information to users in a clear and unobtrusive way. Hence, ubiquitous display solutions are required to support flexible environments, where digital content can be easily accessed under various settings in a given location whilst minimising any feelings of information overload. This chapter starts by briefly addressing the importance of visual information in today's societies and advances on discussing the main printed display technologies currently available, highlighting their suitability for presenting dynamic visual information.

The use of images and symbols as a mean to communicate ideas, beliefs and stories dates back as far as prehistoric times[9]. The amazing images left in the form of cave paintings by earliest known specimens of Homo sapiens (Figure 3-1) mark the very beginning of visual communication. They represent a precious record of a past era.

Since that period in time, Man has searched and experimented various ways to express himself and to communicate with others. From clay or stone sculptures that portray specific feelings and beliefs to carvings and paintings that convey unique moments. The use of ideographic and early mnemonic symbols to visually represent specific information was also common in these yearly times.

[9] For a comprehensive overview of the evolution of visual communication and the history of writing, see (Meggs and Purvis, 2011) and (Robinson, 2007) respectivelly.

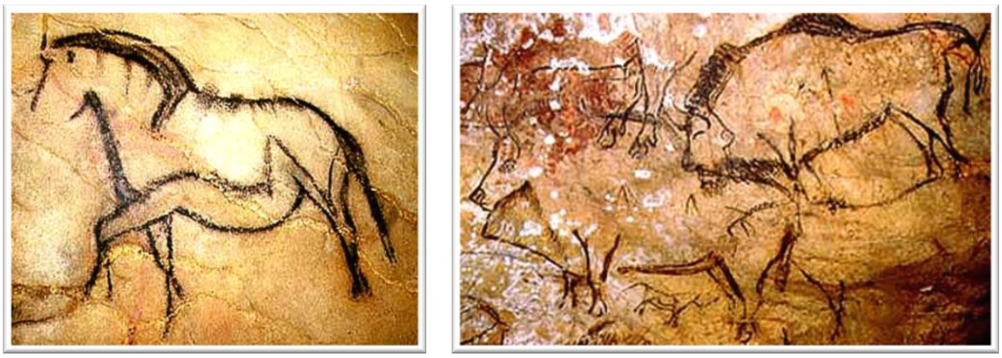

Figure 3-1: Examples of cave paintings. Source: (Clottes, 2002).

With the development of a complete writing system in the Early Bronze Age by the Sumerian (the cuneiform script), human societies gain the means to preserve, systemise and transmit hard-won knowledge, experiences and thoughts in a truly accurate and unambiguous way. The written abstract signs came to represent the sound of the objects depicted instead of the objects itself (i.e. pictures were used as phonograms), making it possible for anyone to reconstruct the exact meaning of writings without having to know the context in advance. The development of the hieroglyphics by the Egyptians represents as well another tipping point in visual communications. Hieroglyphs were used by Egyptians not only to tell the story of their culture, but did do so in a graceful and beautiful visual way. They were the first people to produce illustrated manuscripts in which writing and aesthetically pleasant pictures were combined to communicate information (Figure 3-2). Along with the accomplishments of Mesopotamia, these innovations triggered the development of the alphabet and graphic communications in Phoenicia and the Greco-Roman world (Meggs and Purvis, 2011). The alphabet led to fewer symbols (for example, the Egyptians used approximately 5000 symbols whilst the Phoenician alphabet was composed by only 22 symbols) and a potentially less restricted writing system (Robinson, 2007).

Figure 3-2: Example of an Egyptian illustrated manuscript (Papyrus of Hunefer). Source: (Meggs and Purvis, 2011).

Throughout the centuries, the use of visual elements to communicate information continued to evolve, accommodating new technologies (for example, the printing press) and forms of communication and expression. In the late 1800s, with the improvements made in the motion picture camera, the idea of creating continuous live action became attainable. By making sequential drawings of a continuing action and projecting them onto a screen at a constant rate, it became possible not only to re-create the movements and actions of any living being but as well as to give life to animated objects. Moreover, it was possible to show feelings and emotions in a figure with which the audience actually could become compassionate about (Thomas and Johnston, 1997). Animated graphic soon became an effective method of entertainment as well as of presenting information.

The advent of computers and the internet further pushed the limits of visual communication into new creative directions. It not only enabled new forms of communication and entertainment but also it made available means and tools that increasingly impacted the traditional design and animation practices, facilitating and speeding the production process (Furniss, 2008). Changes of layout, typography or any other graphic element can now be instantly seen without the need of printing, and endless versions of the same work can be easily made. New procedures related to image manipulation and 3D image creation also became available at processing times that could not be done before. The internet on the other hand, provided an easily accessible and affordable medium for exhibition and dissemination of contents with today's websites often integrating multiple disciplines of information systems, information technology and communication design.

The clarity and power that visual elements can offer makes today visual communication a method of choice for conveying information. For example, signs, signage systems, posters and maps are widely used in public spaces such as in airports, train stations, hospitals, museums, schools, or libraries to provide visual directions and instructions. Or, icons and symbols are implemented in vehicles, household appliances, and computers to offer visual clues about specific operating functions.

3.2 Electronic “paper” displays

The compelling need to present information as text and images has led to the development of many different electronic display technologies. The range of options is already high, with each having their own advantages and disadvantages. If we consider the currently available electronic display technologies, it is possible to divide them, according to their lighting source, into two distinct classes: emissive and non-emissive electronic displays (see for instance, (Kahn and Zervos, 2008; Lee et al., 2008)).

In emissive displays, each picture element has the capability to emit its own light. They convert electrical energy into light that serves to illuminate the display screen and form the image. This type of displays perform very well in environments where the luminosity can be controlled or under dark conditions, but tend to lose its viewability, either partially or totally in very bright conditions such as under direct sunlight, where the reflection of ambient light matches or exceeds the light emission from the display (Amundson, 2005; Lee et al., 2008). Examples of emissive devices and technologies include the cathode ray tube displays, field emission displays, plasma displays, electroluminescent displays, and organic light emitting diode displays.

In contrast, non-emissive displays do not emit light and, instead, use optical effects to convert sunlight or light from some external source into graphic patterns. Each picture element operates as an independent light switch, modulating incident light. If the light source is behind the display panel (i.e. a backlight unit is used), the display operates in transmissive mode. Alternatively, ambient light can be used as the light source, and the display is said to operate in reflective mode. Since no backlight is needed in reflective displays, the power consumption of these displays is relatively low (Lee et al., 2008). In addition, novel non-emissive reflective displays are bistable, i.e. their images remain in a non-power-consuming state between image updates. Hence, they only consume energy when the image is being switched and not while it is being displayed. For applications where images are updated only occasionally, the power savings are considerable. Naturally, these displays cannot be viewed in the darkness without an external light source, any more than a book or a newspaper.

Liquid crystal displays (LCD) are currently the dominant technology in non-emissive displays. However, common liquid crystals displays operate in transmissive or transflective mode (Figure 3-3) and thus require a backlight. As a result, like emissive displays, LCDs require a significant amount of energy to generate their own light. Furthermore, they also suffer from the same problem of emissive displays when viewed under direct sunlight or bright light: the image appears washed out. The need for low power consumption displays with a high reflectivity and contrast has increasingly drawn the attention of the research community and led to a quest for alternative display solutions. Research in reflective displays gained momentum and new ideas and technologies have been explored in an attempted to develop paper-like displays.

Novel non-emissive displays hold the promise of being lighter, thinner, more flexible, more adaptable, and more power efficient that any currently available emissive or backlit display. Moreover, by being able to be manufactured using conventional printing technologies, they portend as well low production costs. Clearly, these characteristics show the potential of these displays to facilitate innovative ways to present and interact with information. They allow the fabrication of dynamic environments, the ubiquitous display environment, where access to digital content is supported by a rich variety of display devices embedded in various artefacts and surfaces. Electrochromic displays are one of the most powerful candidates for this purpose. The technology has various merits such as high contrast, low power consumption, optical memory, easy fabrication, substrate versatility, and possibility of transparent and multicolour displays. In addition, a large number of electrochromic materials are available from nearly all branches of synthetic chemistry, widening even more the possible characteristics of electrochromic displays.

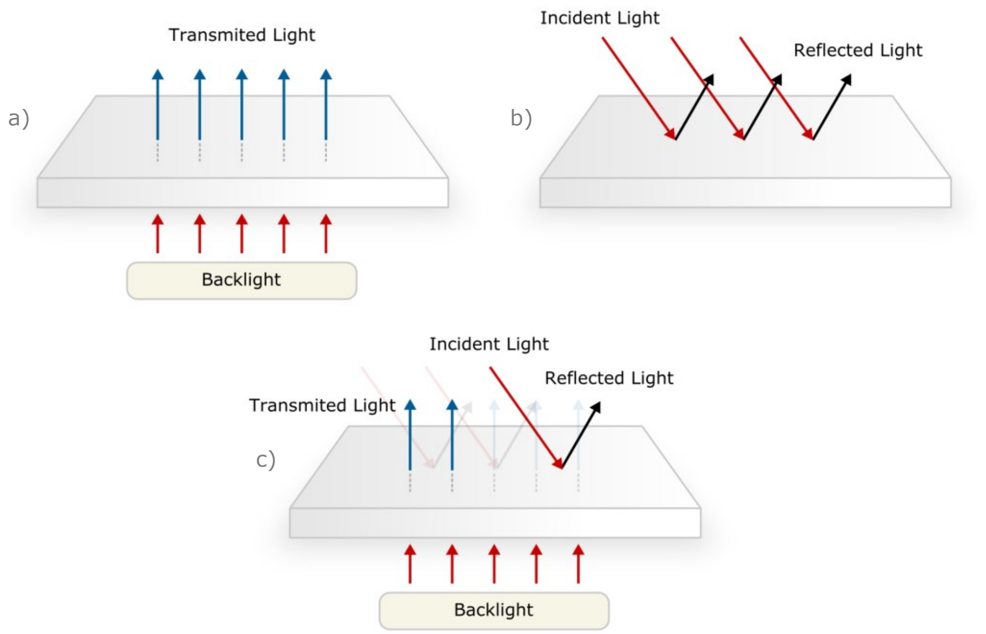

Figure 3-3: Non-emissive displays operation modes: a) transmissive, b) reflective, and c) transflective.

Examples of non-emissive reflective displays include, but are not limited to, electrophoretic displays, electrochromic displays, thermochromic displays, electrowetting displays and photonic crystals displays. In the following subsections it is presented a review of these emergent non-emissive display technologies, describing their functioning, main advantages, challenges, and current (or possible) applications, with a special focus on electrochromic devices. All the technologies reviewed aspire to a certain extend reproduce the visual experience of conventional printed media as seen in books and newspapers. A comparison between the various technologies is provided in section 3.3.

3.2.1 Electrochromic displays

A material is considered to be electrochromic when it exhibits a perceptible colour change in the visible region of the electromagnetic spectrum as a result of an electrical stimulus[10]. This functions by a reversible electrochemical redox reaction

[10] Although this is the typical working definition of electrochromism, and the one adopted for the purpose of this thesis, research in electrochromic devices for multispectral energy modulation has extended it to include also modulation of radiation in the infrared (see for instance, (Franke et al., 2000; Topart and Hourquebie, 1999)) and microwave (see (Rose et al., 1997)) regions. Hence, under this new definition, "colour" can mean a response of detectors to these electromagnetic regions, and not just by the human eye (Rowley and Mortimer, 2002).

that changes the light reflecting properties of the material and hence, alters its perceived colour (see (Granqvist, 1995; Monk et al., 2007)). Electrochromic devices (ECDs) take advantage of this ability by using electrochromic materials capable to change their optical properties in a persistent and reversible way when exposed to an appropriate electrical potential as imaging elements.

Electrochromic devices are typically assembled in a laminate configuration based on a simple two electrode configuration (see Figure 3-4). Two electroactive materials, where at least one is electrochromic, are sandwiched between two electrical conductors (the working electrode and the charge-balancing counter electrode), and separated from each other by an ion-conducting layer that can be a solid, semi-solid or liquid electrolyte. The resulting five layers are commonly protected by two plastic or glass substrates. The application of an appropriate voltage potential to the electrodes results in the migration of electrons and ions, and hence in the redox reaction that causes the colour change in the electrochromic material. The new redox state, and consequently the new colour, persist with little or no input power in the called "memory effect" of the electrolytic cell. By reversing the polarity of the voltage, the ions and associated electrons return to the original layer and the electrochromic material changes back to its original colour. The colour change is commonly between a transparent ("bleached") state and a coloured state, or between two coloured states. When more than two redox states are electrochemically available, the electrochromic material may be able to exhibit several colours (depending on the applied voltage) and can be described as polyelectrochromic (Monk et al., 2007; Mortimer, 1997). Depending on whether the electrochromic material shows colour variation when it is oxidised or reduced, it can be said, respectively, to be an anodically or cathodically colouring material. The molecular region of the electrochromic material capable of impairing a colour is termed chromophore.

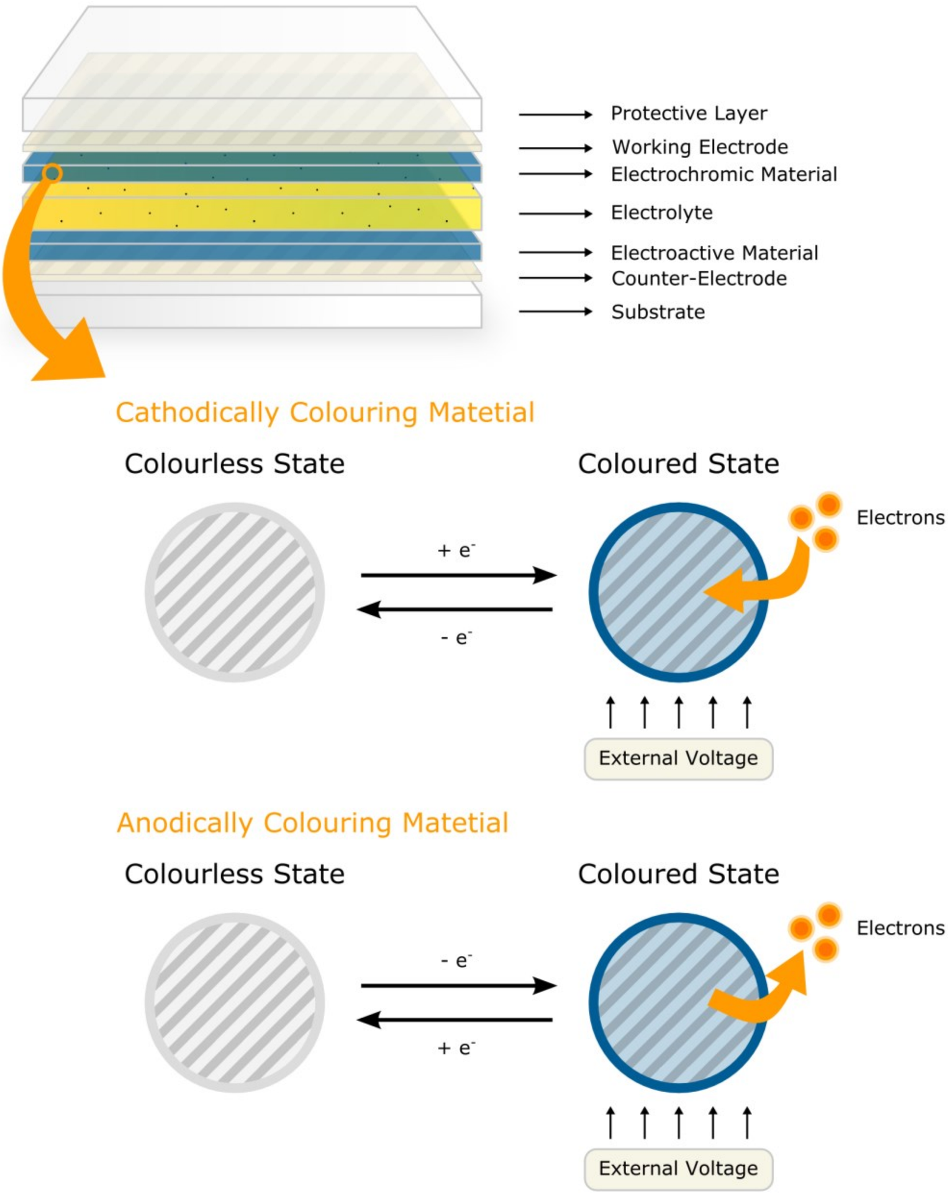

Figure 3-4: Schematic configuration and operating principle of an electrochromic display.

Electrochromic displays can be tailored to display both static (simple) information and dynamic (complex) information. In simple electrochromic displays, a single pre-defined image is patterned on the electrochromic layer, and by changing the colour state of the electrochromic material, the image is either shown or erased (Figure 3-5). The implementation of addressing schemes (see next chapter) where individual picture elements or individual sections of the display can be switched on or off independently, allows the creation of more complex displays capable of producing images dynamically on demand.

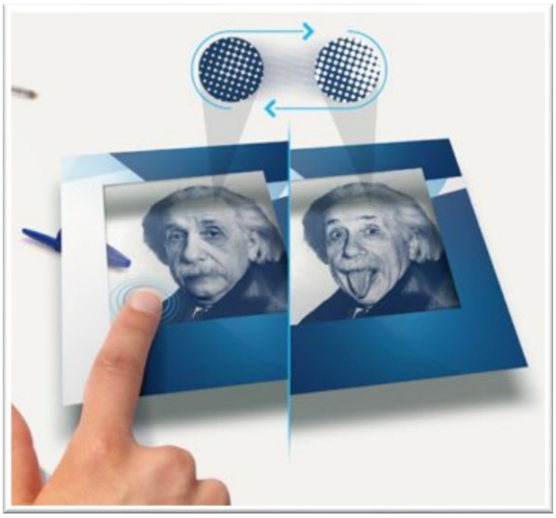

Figure 3-5: Representation of a simple electrochromic display. Source: (Ynvisible, 2011).

The integration of multiple colours, as mentioned, is possible though strongly dependent on the electrochromic material used. Conjugated conducting polymers (see Annex A) are commonly used for this purpose. Various conjugated polymers have more than two redox states and hence, it is possible to change their colour according to different voltage potentials (see for instance, (Ouyang et al., 2011; Tarkuc et al., 2010; Xu et al., 2012)). Moreover, by introducing variations in the polymeric structure, whether by changing the metal species or by modifying the organic ligands used to synthesize the polymers, it is possible to tailor the optical properties of certain conjugated polymers so that other colours are available. The use of two complementary conjugated polymers covering different colour regions, being one cathodically colouring and the other anodically colouring, is also a way to achieve, to a certain extent, multicolour displays (see (Huang and Ho, 2006; Kim et al., 1996)). As colour filters are not used, the high contrast ratio of the displays is not lost.

Another possibility reported in literature, and clearly the most attractive despite being the most complex to implement, consists of obtaining multiple colour representations through an additive (Sonmez, Shen, et al., 2004; Sonmez, Sonmez, et al., 2004) or subtractive (Watanabe et al., 2012; Yashiro et al., 2011) colour-mixture process. This requires three different conjugated conducting polymers, each one capable to generate one of the three additive or subtractive primary colours (red, green and blue, or cyan, magenta and yellow, respectively) from a transparent discoloured status. The intermediate colours are achieved by staking the three primary colours and combining then in the appropriate proportions. The display is assembled in a multi-layered structure divided into a frontplane and backplane (see, (Yashiro et al., 2011)). The frontplane is formed by three superimposed transparent electrodes, each having an electrochromic layer associated to, and separated by insulating layers; and one white reflecting layer. Each electrochromic layer is based on three kinds of organic electrochromic compounds capable to generate one of the three primary colours. The backplane consist of the counter-electrode and is linked to the frontplane through the electrolyte. The structure reduces the light loss to reproduce colour by superimposing the three primary colours, likewise it is done with printing colour on paper.

The main advantages of electrochromic displays are their low power consumption and low manufacture cost. The memory effect of the electrolytic cell ensures that electrochromic displays only require energy when switching between images (i.e. they are bistable). The nature and availability of the raw materials used to produce this type of displays, along with the simplicity of the manufacture process, ensure the low production cost. In addition, electrochromic displays are characterised for having a high reflectivity and a high contrast, presenting the same agreeable readability as printed paper, whether in direct sunlight or in dimmed light.

The main disadvantages of this technology are the lack of resolution of other commercially available display technologies, the limited variety of available colours in large scale production, and the slow response times. As a consequence, electrochromic displays are more suitable for applications requiring simple displays that involve long-term display of information, such as re-usable price labels, advertising billboards and digital signage; or for being incorporated in disposable devices such as smart cards, greeting cards or product packages. Whilst there are many proposed applications for electrochromic devices (see for instance, (Bamfield and Hutchings, 2010; Monk et al., 2007)), until now the most successful commercial applications have been limited to automatically dimming, anti-glare automobile rear-view mirrors (see for instance, (Baucke, 1987; Liu and Richardson, 2005; Lynam, 1987)) and electrically shaded smart-windows (see (Baetens et al., 2010; Cinnsealach et al., 1998; Granqvist et al., 1997; Heuer et al., 2002; Niklasson and Granqvist, 2007; Rauh, 1999)). Both applications are reported in a great number of patents.

3.2.2 Electrophoretic displays

An electrophoretic display is a reflective display that functions based on the electro-static rearrangement of charged, light-scattering, pigment particles suspended in a dielectric fluid, in response to an externally applied electric field. This suspension, commonly called “electronic ink” or “electrophoretic ink”, is the key component of electrophoretic displays.

The first electrophoretic display was reported in 1973, by Ota *et al.* (1973), however, it was only in the late nineties that research in electrophoretic displays boomed. Comiskey *et al.* (1998) demonstrated the first example of a microencapsulated electrophoretic displays, and subsequent work developed by E Ink Corporation[11] led to dramatic improvements in the technology (see for instance, (Albert et al., 2000, 2001; Jacobson et al., 2001; Loxley and Comiskey, 2001)), and to the first products being commercialised (E Ink, 2004). Nowadays, E Ink Corporation continues to be the reference producer of electrophoretic displays.

In E Ink’s electronic ink (Chen et al., 2003; E Ink, 2012g), millions of microcapsules, each about a micrometer in diameter, are suspended in a carrier solution of hydrocarbons and black dye. Each microcapsule contains positively charged white particles (usually titanium dioxide particles) and negatively charged black particles (usually carbon black) in a clear fluid. The basic operating principle of this two-particle microencapsulated system is illustrated in Figure 3-6. When a positive electric field is applied to the bottom surface of a microcapsule, the positively charged white particles are driven to the top of the microcapsule, where they become visible to the user. At the same time, an opposite electric field pulls down the negatively charged black particles. As the particles tend to form a solid layer across the face of the microcapsule, the particles at the top hide the particles at the bottom. Hence, the black particles become concealed to the user. At the top, the titanium dioxide particles reflect the incident light in all directions, and the surface of the display appears white at that point. By reversing the voltage, the black particles migrate to the top of the microcapsule and the white to the bottom. As a result, the surface of the display at that point appears now dark, since the incident light is absorbed by the black dye. By dividing the display into a number of small picture elements, i.e. into pixels, different precise points of its surface can be addressed and manipulated independently. Images can them be easily formed by applying the appropriate electrical charge to each pixel and thus create a pattern of reflecting and absorbing regions.

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[11] http://www.eink.com/

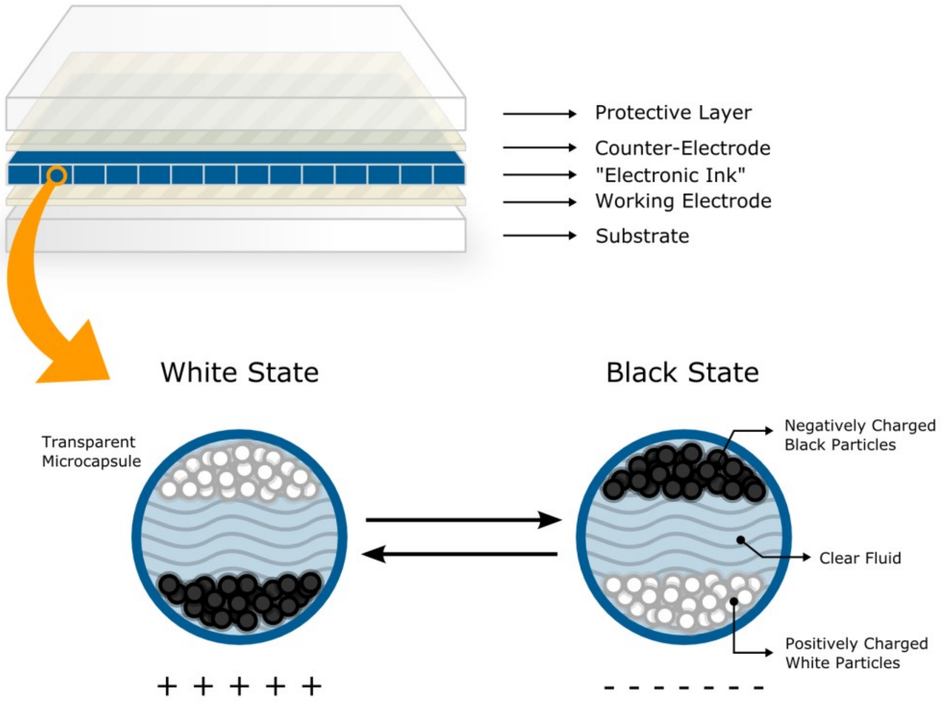

Figure 3-6: Schematic illustration of the principle behind electrophoretic display technology. Source: Based on (Chen et al., 2003).

Electrophoretic displays are characterised for having high white state reflectivity and contrast, and "paper-like" optics. As electronic ink uses basically the same pigments as the ones present in regular ink for books and newspapers, electrophoretic displays have the same agreeable readability and legibility as printed paper. The optical performance of these displays is such that they are insensitive to the intensity and direction of ambient light, possessing excellent readability either in direct sunlight or in dimmed light, under all angles (electrophoretic displays have a viewing angle of almost 180°) (Duthaler et al., 2002). The optical properties of electrophoretic displays are mainly determined by the electronic ink composition. Relevant factors include composition, size, density, and light scattering properties of the pigment particles, as well as concentration of the different compounds (dye and pigments) as a function of the overall thickness of the display (Badila et al., 2008; Werts et al., 2008). In general, a high image quality (high reflectivity and contrast) requires very small particle size with a narrow size distribution. The pigments in the medium must also exhibit good dispersion and stability (Li et al., 2011). Hence, most of the research on electrophoretic displays has been focused on the synthesis of the pigment particles, with the aim of improving the same in terms of scattering properties, surface charges, steric stabilization, interactions with the electrode surface, and inter-particle interactions (see for instance, (Badila et al., 2008; Fang et al., 2009; Jang et al., 2005; Werts et al., 2008)).

The power consumption of electrophoretic displays is also low, mainly due to the long-term image stability and the fact that they do not need a backlight (the main consumer of energy in most displays). In practice, the display only consumes energy when the image is changing. Hence, an image on an electrophoretic display is retained even when all energy sources are removed (E Ink, 2012g). Furthermore, due to the advances witness in the technology of polymer transistors, all components of an electrophoretic display can be made flexible. Hence, it is possible to make bendable or even foldable electrophoretic displays. The slimness of electrophoretic displays is also very close to that of real paper. Indeed, due to the paper like appearance characteristics, electrophoretic displays are usually referred to as electronic paper (e-paper)[12]. However, it must be pointed that there are also other types of non-emissive displays that, by their characteristics can also be categorised as e-paper (namely the ones described herein).

The inherent lack of full colour in electrophoretic displays is commonly presented as one of their major drawback. The functioning principle behind electrophoretic displays enables them only to display shades of colour between two specific colours. As a result, electrophoretic displays are typically monochromatic. Black and white pigment particles are usually used, though other colours combinations are also possible. For instance, Kim et al. (2005), demonstrated the preparation of electrophoretic microcapsules containing cyan and white, magenta and white, and yellow and white pigments. As for full colour electrophoretic displays, these still can be made by using sub-pixelation, i.e. the division of each pixel in the display into sub-pixels, and the addition of a red-green-blue colour filter array over each three sub-pixels (see (Duthaler et al., 2002)). Naturally, this reduces the display brightness by a factor of three as each pixel is limited to a single primary colour and thus, when it is necessary to create a colour in the display, only one-third of the pixels can be used. Another drawback of electrophoretic displays is their low refresh rates, making them, for now, unsuitable for video applications or for sophisticated interactive applications such as scrolling.

[12] E-paper can be used to refer any type of electronic display technology that reproduces the appearance and behaviour of normal paper. E-paper reflects ambient light like any ordinary paper rather than emitting its own light, can be read in direct sunlight without the image appearing to fade, is capable of holding text and images indefinitely without consuming energy (i.e. is bistable), and is slim and flexible like regular paper.

Successful examples of commercial applications of electrophoretic displays include the high resolution active matrix display used in most known e-readers such as the Amazon Kindle, the Barnes and Noble Nook, or the Sony Reader (E Ink, 2012a); or the 10-bar segmented display used in the Lexar JumpDrive Secure II Plus (E Ink, 2012b) that allows customers to easily monitor their USB drive storage capacity. Other examples include various wristwatches (E Ink, 2012e) and mobile phones, such as the Motorola Motofone F3 (E Ink, 2012d) whose main screen is an electrophoretic displays. Figure 3-7 illustrates some of these examples.

Figure 3-7: Examples of commercially available applications of electrophoretic displays. Source: (E Ink, 2012b, 2012c, 2012f).

SiPix Imaging[13] is another reference company in the field of electrophoretic displays. Their approach to electrophoretic displays is different from E Ink in the sense that they use a different method for encapsulating the charged particles and a different type of electrophoretic fluid. In SiPix's electrophoretic displays, the electrophoretic fluid is composed only by electrically charged white micro-particles, which are dispersed in a coloured dielectric solvent, as opposite to a clear one. Hence, while the white particles ensure the white state, the coloured solvent guarantees the black (or coloured) state. Moreover, the electrophoretic fluid is enclosed in a three-dimensional embossed matrix structure, named by SiPix of Microcup structure, rather than in microcapsules (Liang, Hou, Zang and Chung, 2003; Liang, Hou, Zang, Chung, et al., 2003).

[13] http://www.sipix.com/

3.2.3 Quick-response liquid powder displays

Quick-response liquid powder displays (QR-LPD) operate in a very similar way to electrophoretic displays. The displays are composed by two types of powder, one with white colour and negative charge, and the other with black colour and positive charge (see, (Hattori et al., 2003, 2004, 2010)). Both types of powder are placed together into an area between two patterned substrates. Separators are used to form cells gaps and prevent the mixing up of powders between adjacent pixels and ensure the uniform distribution of the powders within each pixel. The rest of the space in the cell is filled with air. The powders are attracted to each other and make a mass with grey colour, yet each powder behaves just like a liquid by itself. It is the extremely high liquidity of the powders that allows the use of the term "Liquid Powder" for this material (i.e. the materials have a powder form but show a liquid behaviour). When a negative voltage is applied to the upper transparent electrode, the positively charged black powder moves to the upper electrode exhibiting a black appearance and, in the opposite bias case, the negatively charged white powder is attracted to the upper electrode exhibiting a white appearance. Due to attraction forces (electrical and non-electrical) between the electrodes and the liquid powder, images can be maintained without electric power (Hattori et al., 2003, 2004, 2010). However, separating the charged powder pigments from each other and from the electrodes requires significant force (unlike in liquid dispersed pigments in electrophoretic displays). As a result, operating voltages are high (around 40V to 70V) (Hattori et al., 2004, 2005; Heikenfeld et al., 2011; Sakurai et al., 2007).

QR-LPD present response times of less than 0.2 msec, bright images with a paper-white appearance with more than 40% in reflectivity, bi-stability, wide viewing angle with nearly ideal diffuse reflectance, large matrix-addressing capability by passive-matrix driving, and halftone images with more than four gray levels (Hattori et al., 2004). Colour displays were also demonstrated using two different methods (Sakurai et al., 2006). The first method consisted of using colour liquid powder to produce coloured monochromatic displays, while the second method consisted of applying colour filter technology (RGB colour filter) with black and white liquid powder to produce a full colour displays (Figure 3-8).

Figure 3-8: Picture of a full-colour quick-response liquid powder display. Source: (Hattori et al., 2010).

QR-LPD technology was first demonstrated in 2003 (Hattori et al., 2003), by Bridgestone Corporation[14]. In 2012, however, Bridgestone announced its plans to withdraw from the electronic paper business and abandon the research on quick-response liquid powder displays (Bridgestone, 2012).

3.2.4 Electrowetting displays

Electrowetting displays employ principles of microfluidic to manipulate the shape of a confined water-oil interface through an applied voltage (see (Hayes and Feenstra, 2003; Pollack et al., 2000)). Each pixel of the display is formed by an optical stack comprising of a white (reflecting) substrate, a hydrophobic insulator, a droplet of coloured oil and water (see Figure 3-9).

In equilibrium, the droplet of coloured oil forms a continuous film between the water and the hydrophobic insulator, resulting in a coloured pixel in the display. However, when an electrical potential is applied across the hydrophobic insulator, the interfacial tension between the water and the hydrophobic insulator changes, and the initial stacked state is no longer energetically favourable. As a result, the system proceeds to lower its energy by moving the water into contact with the insulator, and thus displacing the oil. The underlying subtract becomes visible, and the optical properties of the stack are changed from a coloured off-state (due to the colour of the oil) to a white reflective on-state (due to the colour of the subtract where the droplet of oil is placed). Still, the size of the pixel must be sufficiently

[14] http://www.bridgestone.com/

small so that the viewer does not see the droplet of oil contracted, and instead experiences the average optical response (Feenstra et al., 2004; Hayes and Feenstra, 2003). When the voltage is removed, the oil droplet returns to is initial state.

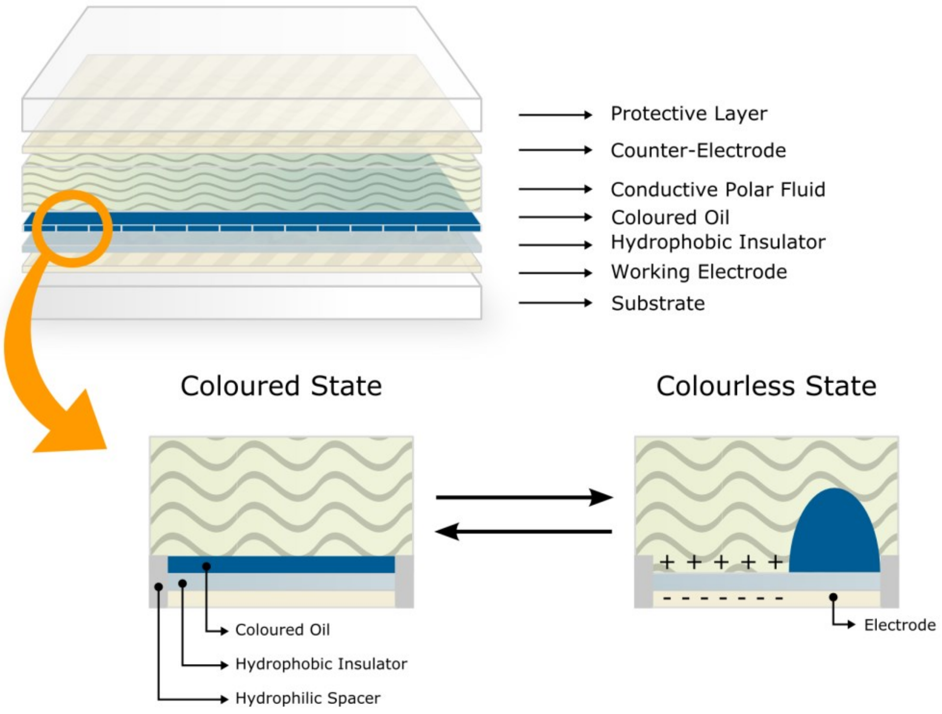

Figure 3-9: Schematic illustration of the principle behind electrowetting display technology. Source: Based on (Feenstra et al., 2004).

Electrowetting technology can be used to produce simple black and white displays, or to produce a wide range of displays with different colours, simply by applying different dyes with different colours (Figure 3-10). Multiple colours displays can be achieved either by using the single layer architecture described above with a black coloured dye and a multiple colour filter (i.e. employing sub-pixelation), or by implementing a three layer architecture, where each layer correspond to a different monochromic colour. In this type of setting, the layers function independently of each other, and are activated by the same oil dosing process that is performed on single layers. Since any colour can be generated anywhere in the display surface, the optical performance increases with a factor of three with respect to the single layer structure.

Figure 3-10: Picture of various electrowetting displays showing diverse colours. Source: (Feenstra and Hayes, 2009).

In addition to displaying colour, electrowetting displays are also capable to show video content (the response time is around 10ms) (Feenstra and Hayes, 2009). In sum, electrowetting technology allows high brightness, high contrast, and full colour displays capable of presenting video content. The technology can also operate in other modes beyond reflective, namely, in transmissive, transparent, and transflective. Their main disadvantage is not being truly bistable.

Possible applications for electrowetting displays include displays for e-readers (depicted in Figure 3-11); signage, billboards and other public displays; low end displays for devices such as digital watches, MP3 players or calculators; and high end displays for laptops and notebooks, tablets, smartphones, automobile navigation systems, digital cameras and camcorders, or any other device currently having an LCD screen. Liquavista[15] is currently the reference company in electrowetting displays.

[15] http://www.liquavista.com/

Figure 3-11: Liquavista's electrowetting display e-reader. Source: (Stevens, 2010).

3.2.5 Interferometric modulation displays

Interferometric modulation (IMOD) displays are reflective displays that have at their core microelectromechanical systems (MEMS)[16]. Each pixel of the displays is composed by a conductive and light reflective membrane, which is separated from a conductive thin-film stack by a thin air gap (Figure 3-12). Depending on the voltage applied (high or low) to the reflective membrane and the thin-film stack, the membrane can be positioned into two distinct positions, each representing a specific state of reflection. A high voltage level results in the membrane experiencing electrostatic attraction and being drawn towards the thin-film stack. As a result, the air gap is closed and the light being reflected is shifted to the ultraviolet spectrum: the pixel appears black. This is called the "collapsed state". In turn, the application of a low voltage level results in the membrane positioning itself far from the thin-film stack. The air gap becomes open and light is reflected in a certain wavelength of colour. The pixel appears bright and coloured. This is called the "open state". The wavelength reflected, and thus the colour exhibit, is determined by the distance between the membrane and the thin-film stack. However, this distance is fixed upon construction of the display and not determined by the voltage potential applied. IMOD displays are designed to operate in a binary way, switching from one colour to another, or from one colour to black (Miles, 1997; Miles et al., 2002).

[16] For an introduction to the principles of microelectromechanical systems, see for instance, (Maluf and Williams, 2004).

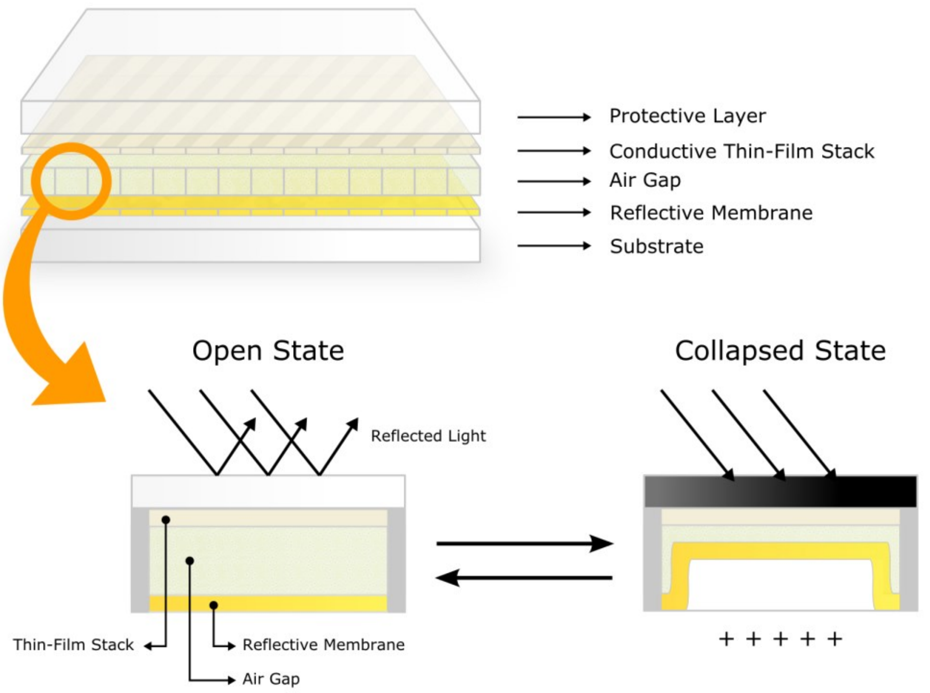

Figure 3-12: Schematic illustration of the functioning principle behind an interferometric modulation display. Source: Based on (Miles, 1997; Miles et al., 2002).

Full colour displays are achieved through the implementation of sets of three IMODs sub-pixels arranged in spatial RGB stripes where each set is capable to individually switch colour from either red to black, green to black, or blue to black. This implementation does not require the use of colour filters, as colour is generated using the sub-pixels air gaps. However, this implies three different air gap thicknesses for each colour sub-pixel. White is achieved by an additive mixture of all three colour stripes (Miles, 1997; Qualcomm, 2008).

The mechanical structure of the IMOD pixel also results in low power consumptions. The technology is bistable, meaning it requires very little power to maintain either of its two states. The electric charge can be manipulated and maintained to the point at which written imagery can persist indefinitely (Miles, 1997; Qualcomm, 2008). Moreover, as the displays are highly reflective, they not only can be used in direct sunlight, but also do not require any extra illumination. The fact that IMOD displays are bistable also results in being passive-matrix addressing. IMOD displays also offer the fastest switching speeds on reflective displays. Each pixel in an IMOD display can change its state in less than 20 microseconds (Miles, 1997). This characteristic makes them suitable for applications that require video capability. However, in full colour IMOD display, due to the side-by-side configuration the reflectance is limited to less than 33%. The viewing angles of the displays, although it can exceeds 60° (Miles et al., 2002), it is also limited due to the nature of the physics behind the colour reflection capability. Heikenfeld *et al.* (Heikenfeld et al., 2011) also points out that the manufacturing cost of IMOD displays is high due to low yields, and that the displays are unlikely to be processed in flexible substrates because of the complexity of the MEMS structure and the use of inorganic dielectrics.

IMOD displays where first reported by Miles (1997), in 1997. The technology was afterwards acquired by Qualcomm[17], which is currently commercialising it under the trademark name mirasol (Qualcomm, 2013a). To date, various e-readers where already produced using the mirasol display (Qualcomm, 2013b). Figure 3-13 illustrates one.

Figure 3-13: Picture of an interferometric modulation display e-reader fabricated by Qualcomm. Source: (Qualcomm, 2013a).

3.2.6 Photonic crystals displays

Photonic crystals (see for instance, (Joannopoulos et al., 2008)) are optical nanostructures, structurally arranged in a regular pattern, capable of affecting the motion of photons. By changing the structural pattern of the crystals, it is possible to change the wavelength of the light being reflected and thus, modify the colour being displayed by the crystal.

[17] http://www.qualcomm.com/

The first full colour displays based on photonic colour were developed by Opalux Inc.[18]. They consisted of an expandable electroactive polymer gel/silica opal composite which could be electrically stimulated. The technology was named Photonic Ink, or simply P-Ink, by Opalux (Arsenault et al., 2007). Performance issues led Opalux to improve the technology, implementing an electroactive inverse polymer-gel opal in which the electrolyte freely infuses the nanoporous lattice (Puzzo et al., 2009).

In its essence, each picture element of a Photonic Ink display is composed by an artificial photonic crystal containing hundreds of 200 nm diameter silica microspheres embedded in a porous electroactive polymer containing iron atoms. This composite film, i.e. the P-Ink film, is placed between a pair of electrodes along with an electrolyte fluid. When a positive voltage is applied to the electrodes, the iron atoms in the polymer can lose electrons and become positively charged. This results in an influx of negatively charged counter-anions from the electrolyte to the electroactive polymer, in order to maintain the electrical neutrality of the P-Ink film. As the electrolyte is drawn into the electroactive polymer, the latter expands. The silica microspheres are pushed apart, changing the refractive index of the photonic crystal and thus its perceived colour. The degree of expansion of the silica microspheres is controlled by the voltage applied (Figure 3-14), and can be reversed by an equivalent negative voltage potential (Wang et al., 2011).

Since it is the inter-sphere spacing that dictates the reflected colour of the material, photonic crystals displays can be manoeuvred to display the entire visible spectrum simply by changing the applied voltage. Hence, each pixel can be individually tuned to any colour. Furthermore, once a pixel has been tuned to a colour, it can hold that colour without consuming any power (Wang et al., 2011). A series of pictures illustrating different colour states of a P-Ink display are shown in Figure 3-15.

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[18] http://opalux.com/

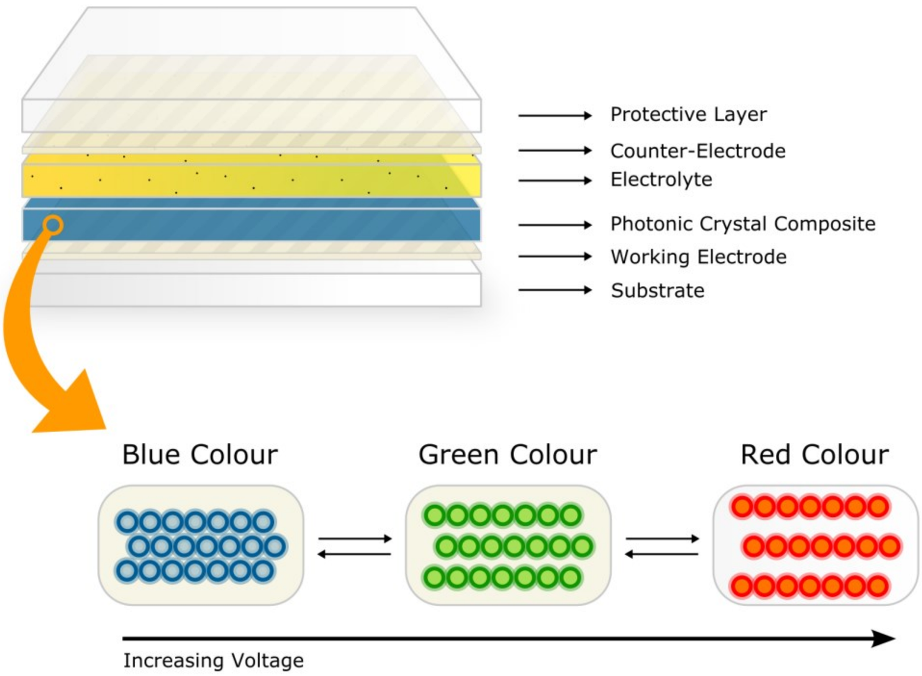


Figure 3-14: Structure of a photonic crystals electrochemical cell showing the effect of film thickness on reflected colours. Source: Based on (Graham-Rowe, 2007; Opalux, 2013).

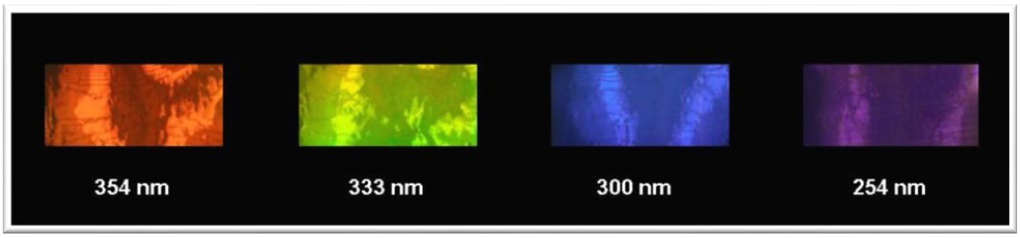


Figure 3-15: Different colour states of a P-Ink display. Source: (Opalux, 2013).

The first displays developed by Opalux, as briefly mentioned, presented some major performance issues, namely, slow switching times (around one second), which were even more pronounced when pixels changed from long wavelengths colours to shorter ones (or vice versa), and narrow viewing angles for each single colour, where each colour was only stable within approximately plus/minus five degrees from certain directions (Ellis, 2007). The implementation of the described structure greatly improved the overall performance of photonic crystals displays. By increasing the specific surface area of the P-Ink film in contact with the electrolyte, electron and ion diffusion lengths were reduced. This resulted not only in increased switching speeds (high enough that switching between any spectral colours in the visible spectrum takes less than 0.2 seconds) but also in a decrease in the driving voltage needed to power the device (Puzzo et al., 2009).

The main advantage of this technology is undoubtedly the capacity to individually control the colour of each pixel without the use of colour filters. However, as Heikenfeld et al. (2011) points out, the technology still needs refinements in terms of the white state. Also, it does not possess inherent grey scale. Nonetheless, the unique capability to tune to any colour of the visible spectrum, along with the low operating voltages (colour switching is carried out at voltages inferior to 1.5 V) and bi-stability, as well as the high reflectivity, high brightness, and fast switching speeds, render photonic crystals displays a viable option for next-generation full-colour reflective displays. Indeed, they show a great potential for a wide range of applications, from e-book readers and flexible mobile displays to large area advertising displays, smart packaging and smart cards. Figure 3-16 illustrates a simple seven-segment photonic crystal display tuned to different colours.

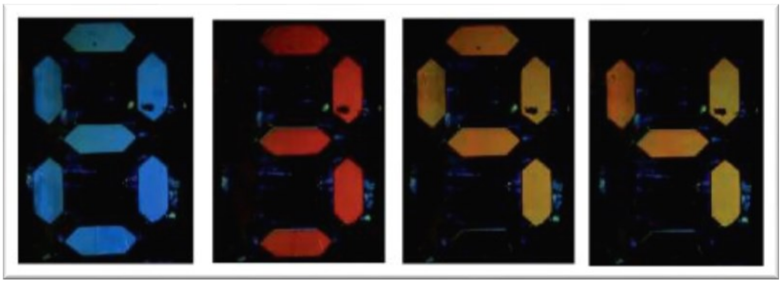


Figure 3-16: Seven-segment P-Ink display demonstration. Source: (Heikenfeld et al., 2011).

3.2.7 Thermochromic displays

Thermochromic displays take advantage of the property that certain elements or materials have to change their colour as a response to specific variations in the temperature. Hence, thermochromic displays function based on temperature-induced colour changes.

Thermochromism can be found in various materials, either organic or inorganic (see for instance, (Day, 1963, 1968; Kiri et al., 2010)). Furthermore, thermochromic systems can display either reversible or irreversible colour changes on heating or cooling, including coloured to colourless, colourless to coloured, and colour to colour changes. Nowadays, the most important thermochromic materials used in commercial products involve either thermochromic liquid crystals or organic leuco dyes (White and LeBlanc, 1999). In general, thermochromic liquid crystals systems are mainly used in applications where temperature accuracy is important (they can be highly temperature sensitive), while thermochromic leuco dyes systems are used where larger temperature differences are present or when temperature accuracy is less important. However, leuco dyes systems are less expensive and more robust than thermochromic liquid crystals systems. They also allow for a wider range of colours and can be used in a wider range of materials. Hence, for display applications, thermochromic leuco dyes systems are often a viable low cost solution.

Thermochromic leuco dyes systems (see for instance, (Burkinshaw et al., 1998; MacLaren and White, 2003; White and LeBlanc, 1999)) typically consist of three components: a colour former (the leuco dye), a colour developer (usually a weak acid), and a solvent (an alcohol or an ester). The three components are mixed together in millions of microcapsules that are printed directly in the surface of the display. As the leuco dye molecules react to changes in the pH by alternating between two states, where one is coloured and other colourless, their interaction with the colour developer defines the colour state of the mixture (Kulčar et al., 2010) (Figure 3-17). Hence, the reversible colouration and discolouration of the thermochromic ink results from the alternation of two competing reactions, one between the leuco dye and the colour developer, and the other between the colour developer and the solvent (Kulčar et al., 2010; Siegel et al., 2009).

The first of these two interactions prevails at lower temperatures, where the solvent exists in its solid form, and the leuco dye and the colour developer form a complex that results in the coloured state of the leuco dye molecules, and therefore, in the colouration of the thermochromic ink. At higher temperatures, where the increase of temperature causes the microcapsules to heat up, and the solvent inside to melt, the second reaction prevails. The solvent reacts with the colour developer, increasing the pH of the solution. The leuco dye and colour developer complexes dissociate, and the leuco dye molecules change to their colourless state. Since this reaction is reversible, when the thermochromic dye colds down, the solvent re-solidifies and the interaction between the leuco dye and the colour developer prevails once again. As a result, the leuco dye molecules return to their original colour state. By mixing the thermochromic ink with traditional dyes and pigments, it becomes possible to create other colour transitions, such as changes between two distinct colours.

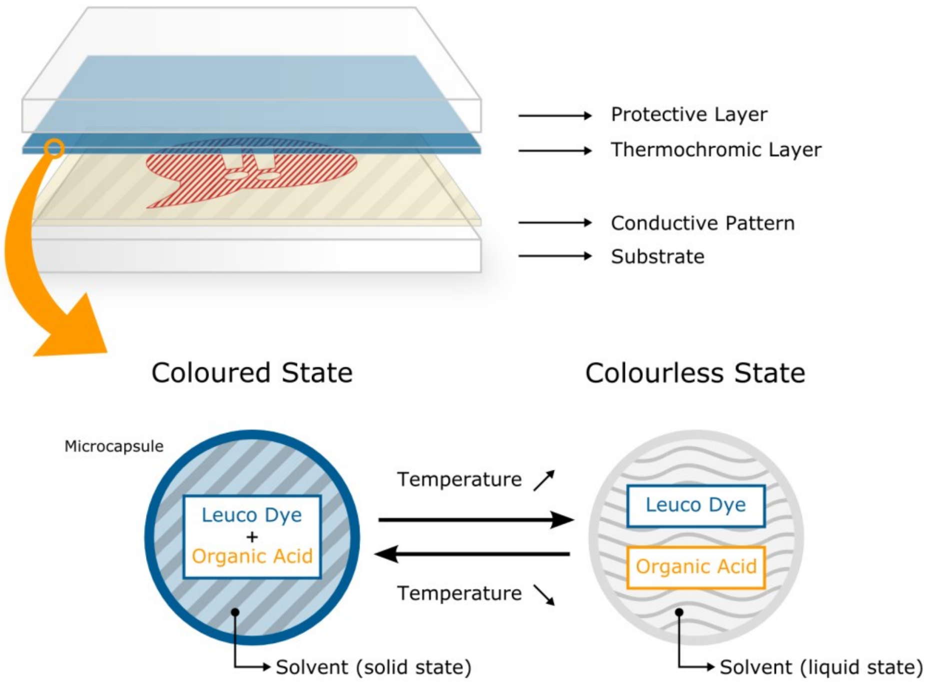


Figure 3-17: Schematic illustration of the operation principle of a leuco dye-based thermochromic display.

Leuco dye-based thermochromic inks are available at various activation temperatures, however most applications are limited to three standard temperature ranges, namely cold (~10ºC), body-heat activated (~31ºC) and warm (~43ºC) (Kulčar et al., 2010). The thermochromic ink is activated by a heat generating system, which can involve simple human touch (Figure 3-18) (see for instance, (Berzina, 2011)), or more complex methods incorporating micro conducting wires and electronic circuitry (Figure 3-19) (see (Liu et al., 2007; Siegel et al., 2009)).

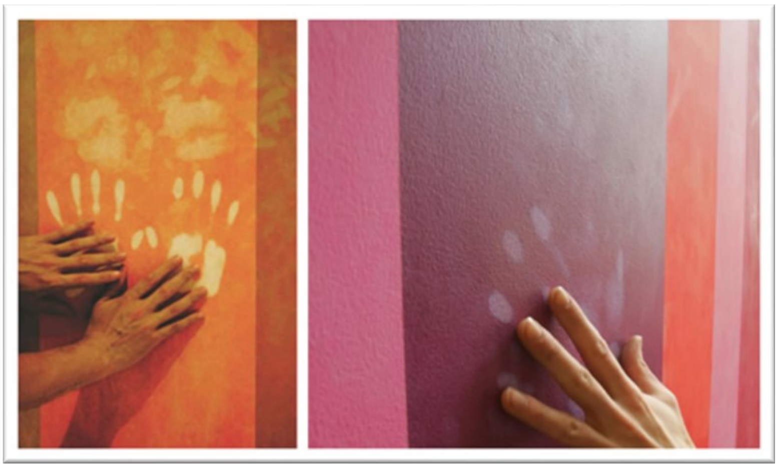


Figure 3-18: Thermochromic wallpaper activated by human touch. Source: (Berzina, 2011).

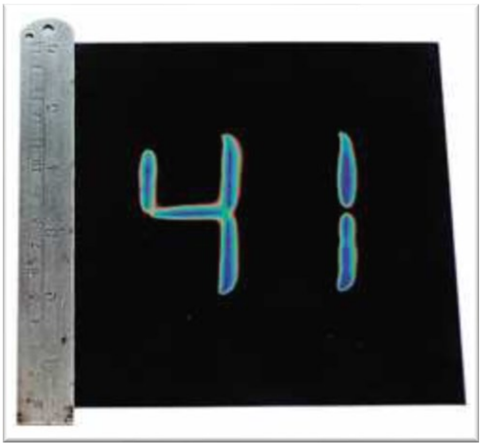


Figure 3-19: Seven-Segment Thermochromic Display. Source: (Ramsey et al., 2000).

When compared to other non-emissive displays, thermochromic leuco dye-based displays present a simpler structure and are cheaper and easier to manufacture. Also, when electricity is used to activate the display, the power consumptions are not dependent on the total surface area of the display, but rather on the power requirement to turn the desired pattern visible (Maas et al., 2009). In addition, as Choi et al. (2010) demonstrated, the power consumption of the display can be successfully minimised by implementing the pulse width modulation (PWM) technique. Their biggest weakness lies in the high switching times, especially when the display is deactivated. In general several seconds are necessary for the temperature to drop to the initial conditions, and therefore, for the colour to revert to its original status. During this period of time, any printed pattern gradually fades away. Hence, thermochromic displays are essentially appropriate for applications that do not require frequent changes in the display content. There are several parameters that can affect the switching times of thermochromic leuco dye-based displays, namely, the voltage applied, the temperature of the ambience, the quality of the paper, and, naturally, the activation temperature of the thermochromic ink (Hennerdal and Berggren, 2011; Liu et al., 2007). For instance, by increasing the voltage applied, faster switching times can be obtained, however, if the duration or amount of the voltage applied is too much, the display can be overheated, leading to blurred images.

Thermochromic displays are commonly used to produce dynamic colours in products, such as mugs, t-shirts, magazines or toys; as temperature sensors when the temperature response accuracy is not critical; as warning messages and safety signs for products reaching certain temperatures; as security element for tickets, smart cards or documents; or as temperature indicators implemented in packaging products, such as beverages, ice-creams, or microwaveable products. Another good example is the charge indicator on Duracell batteries. For more complex applications, the implementation of specifically designed electronic circuitry, with the aim to provide controlled and regulated temperature profiles to specific areas of the display, enables the production of dynamic information, in opposite to displaying only pre-programmed images. For instance, Glaister and co-workers (Glaister et al., 2004) incorporated thermochromic materials into concrete, in combination with a system of nickel-chromium wires connected to a power source, in order to show low content information at the surface of the concrete. The implementation of matrix addressing schemes, composed by thermal pixels, has also been demonstrated (Yarimaga et al., 2010).

3.3 Technology comparison

This section attempts to highlight the main differences between the various non-emissive display technologies previously addressed. In Table 3-1 it is provided a side-by-side comparison between the different technologies based on key performance factors. Both visual and operating performances are analysed, as well as current state of maturity for each technology.

All the technologies considered are characterised for presenting paper-like optics. They all possess a white state reflectivity higher than 40%[19] and a contrast

[19] Note that the surface of printed newspaper has a maximum reflectivity of 62% (Hattori et al., 2004).

ratio in the order of 10:1. The viewing angle of all displays is as well high. With the exception of photonic crystal displays, all the technologies have a viewing angle higher than 60º. Also, since they are all non-emissive display technologies, they all can be used in direct sunlight without the image appearing to fade. They are as well, with the exception of IMOD displays, flexible to the point of mimic regular paper. As for the power consumption, it is considered low in all the display technologies considered, with most of them being capable to hold text and images without consuming energy for at least a few hours. The exceptions are electrowetting displays and thermochromic displays which are not truly bistable.

An analysis focused on the "Operation Performances" of the various display technologies promptly highlights three of the technologies by their capability do show video content. These technologies are QR-LP displays, electrowetting displays, and IMOD displays. They all present switching times below 10 msec, with IMOD displays presenting the lowest switching times, in the order of 0.01 msec. None of the other technologies considered presents switching times of this order. In addition to being capable to display video content these technologies are also capable to display full-colour. Their biggest drawback resides in their power consumptions or required driving voltage. IMOD displays have the highest power consumption of the technologies considered here (excluding thermochromic displays), and QR-LP displays require the highest driving voltages to operate. Electrowetting displays present performances between both, being that, as mentioned, they are not bistable. The manufacturing complexity and production cost of these technologies also, already led to the companies responsible for the production of two of these technologies to rethink their strategies. Bridgestone announced their intentions to completely abandon the production of QR-LP displays (Bridgestone, 2012), and in a comparable way, Qualcomm announced their plans to limit the production of mirasol displays (IMOD technology) on a set of products and try to license the technology to other companies (EE Times, 2012). It must be pointed, nonetheless, that these specific technologies intended to position themselves as direct competitors of current LCD technology. The superior performance of these technologies makes them not only suitable for use in mobile applications such as e-readers, mobile phones, portable media players and digital cameras, but also appropriate, if the technology can be successfully scalable, to be used in high end products such as televisions and laptops. The three technologies have already been successfully demonstrated or, in the case of IMOD display technology, commercialised in several colour e-readers.

Table 3-1: Side-by-side comparison of different non-emissive display technologies based on various performance factors.

	Electrochromic Displays	Electrophoretic Displays	QR-LP Displays	Electrowetting Displays	IMOD Displays	Photonic Crystals Displays	Thermochromic Displays
Visual Performances							
Contrast Ratio	High (not specified)	10:1	10:1	15:1	10:1	High (not specified)	N/A
Reflectivity (%)	> 60	> 40	40	60	50	60	N/A
Greyscale Level	4-bit display demonstrated	4-bit	4-bit	4-bit	3-bit	No	N/A
Colour Approach	Multiple monochromatic colours; Full-colour demonstrated with 3-layer architecture.	Multiple monochromatic colours; Full-colour with sub-pixelation and RGB filter.	Multiple monochromatic colours; Full-colour with sub-pixelation and RGB filter.	Multiple monochromatic colours; Full-colour with 3-layer architecture.	Multiple monochromatic colours; Full-colour with sub-pixelation (RGB stripes).	Photonic Colour (each pixels is capable of displaying the entire visible spectrum)	Multiple monochromatic colours; Possibility of thermal pixels with RGB colours.
Viewing Angle	~ 180º	~ 180º	Wide (not specified)	70º	60º	30º	> 80º
Substrate Flexibility	Flexible	Flexible	Flexible	Flexible	Rigid	Flexible	Flexible
Screen Resolution reported	4.8 inch display with 100 ppi	13.3 inch display with 300 ppi	21 inch display with ~ 76 ppi	6.3 inch display with 167 ppi	5.7 inch display with 223 ppi	N/A	N/A

(Continued on next page)

	Electrochromic Displays	Electrophoretic Displays	QR-LP Displays	Electrowetting Displays	IMOD Displays	Photonic Crystals Displays	Thermochromic Displays
Operation Performances							
Driving Voltage (V)	1 to 5	15	40 to 70	15 to 20	~ 10	< 1.5	5 to 15
Bistable	Yes (~ 2 to 3 h)	Yes	Yes (months)	No	Yes (> 10 h)	Yes (hours)	No
Power Consumption (when switching image) (mW/cm^2)	< 3	~ 1.5	< 0.5	< 4.5	< 15.5	Low (Not specified)	~ 50
Switching Time (msec)	100 to 1000	120 (up to 980 in colour mode)	0.2	< 10	0.01	200	> 1000
Video Content	No	No	Yes	Yes	Yes	No	No
Matrix Drive	Passive-Matrix Active-Matrix	Active-Matrix	Passive-Matrix	Active-Matrix	Passive-Matrix	PM / AM not demonstrated	Active-Matrix (thermal pixels)
Stage of Development	Demonstrated in segmented and matrix addressed displays; Commercialised in products, such as smart windows or postcards.	Commercialised in several products, from e-readers to wristwatches.	Demonstrated in segmented and passive-matrix displays (e.g. e-readers from Delta Electronics).	Demonstrated in several e-readers prototypes.	Commercialised in e-readers (e.g. KYOBO e-reader).	Demonstrated in segmented displays.	Commercialised in several products, from t-shirts and mugs to temperature sensors and security elements.
References	(Chung et al., 2010; Pettersson et al., 2004; Sonmez, Sonmez, et al., 2004; Yashiro et al., 2011; Ynvisible, 2012)	(Duthaler et al., 2002; E Ink, n.d., n.d., n.d.; Heikenfeld, 2011; Pitt et al., 2002)	(Hattori et al., 2004, 2005; Masuda et al., 2006; Sakurai et al., 2006, 2007)	(Feenstra and Hayes, 2009; Feenstra, 2006; Hayes et al., 2004; Heikenfeld, 2011)	(Miles et al., 2002; Qualcomm, 2008, 2013b)	(Opalux, 2013; Wang et al., 2011)	(Hennerdal and Berggren, 2011; Liu et al., 2007; Siegel et al., 2009)

From all the display technologies considered, thermochromic displays (leuco dye-based) are the ones that present the worst performances either in terms of power consumption, required driving voltage, and switching time. Their main attractiveness is mainly related to the easiness with which they can be manufactured and the aptitude to directly created the display in almost any type of substrate or product. This is the main reason why this type of technology is widely used to produce dynamic colours in products, such as mugs, t-shirts, magazines or toys. Nonetheless, this does not mean this type of technology can only be used for this type of purpose. As already mentioned in section 3.2.7, thermochromic displays can also be used as dynamic information displays in various applications, from temperature indicators implemented in packaging products to segmented displays embed in concrete.

The remaining three display technologies (electrochromic displays, electrophoretic displays and photonic crystal displays) present similar operating performances, although each one is at a different stage of maturity. The power consumptions of the three technologies are relatively low, with electrophoretic displays having the lowest power consumptions of the three. The driving voltages are as well low, mainly in electrochromic and photonic crystals displays. Also, even though these three technologies are not capable of showing video content, their switching time can be as low as 200 msec. Electrophoretic display technology is currently the technology with the best proven track record, even when compared to the others technologies considered in this analyse. It is a well-established and well-proven technology being commercialised in several products, from e-readers to wristwatches, already for some years. Indeed an attractive choice for display devices where video content is not relevant. Photonic crystal display technology, on the other hand, is in its infancy. Until now, the technology has only been demonstrated in single cell displays and segmented displays. Despite possessing one of the most interesting features of all the display technologies highlighted here, the ability to individually tune each picture element to a specific colour of the visible spectrum, photonic crystal technology still requires considerable development in order to become commercially viable. As for electrochromic displays, the technology is being steadily studied since the early seventies and is currently used in commercial products such as smart windows, rear-view mirrors, or greetings postcards. Noteworthy that commercialisation is mainly focused on monochromatic displays, with multiple colour or full-colour displays still in the lab stage. The attractiveness of this technology resides essentially in the relation between the visual and operation performances associated to a low production cost. As the manufacturing process of electrochromic displays is relatively simple and relies essentially on common printing techniques, such as screen and inkjet printing, the cost of production is largely reduced when compared to other display technologies. These characteristics make this technology particularly suitable for use as widespread low tech information displays. In sum, in applications where cost and power consumption must be low, the switching speeds fast enough to allow quick updates in the content of the displays but not necessarily fast to show video content, and high visual performances are required but full-colour is not fundamental (for instance, in electronic labels and electronic signage).

Paper is considered the display technology by excellence. It is being used already for two millenniums and it still continues to have a fundamental role in the presentation and divulgation of information. However, its content, by nature, is static. Electrochromic displays, by mimicking certain properties of regular paper, such as the look and agreeable readability, with the capacity to display dynamic information can combine the best of the two realities and enable new forms for users to consume information.

3.4 Concluding remarks

Visual information has an important role in today's societies, being used practically everywhere, from physical formats such as road signs and retail displays to digital formats such as websites. The ability to transform traditionally passive physical formats of visual information into active formats capable of presenting information that is changeable and updatable in response to an external stimulus opens new opportunities for presenting information. Inevitably, these displays will have to have a low production cost, low power consumption, be flexible, lightweight and robust, as well as viewable under various light conditions, including under direct sunlight. This section provided an overview of currently emerging printed non-emissive electronic display technologies. It described the technical breakthroughs behind each type of technology, highlighting its advantages and limitations. Furthermore, it presented existing and possible future applications.

The technologies reviewed allow the development of cheap, low power, ultra-thin flexible displays with a high reflectivity and contrast. Due to these characteristics, they bear the promise of revolutionising the display market. From this review, it was possible to verify that non-emissive display technologies are already competing for certain market areas with emissive displays and backlit liquid crystal displays. This is particularly visible in low end electronic devices where non-emissive displays are starting to be seen as an economic and ecological alternative. In the high end segment, considering that technologies such as plasma displays and liquid crystal displays are reaching their economic limit in the size, constructors are also starting to see non-emissive displays as an alternative. However, before non-emissive electronic display technologies can be truly successful, some improvements are still required. First, non-emissive electronic displays have to become capable to display high resolution colours while maintaining the high contrast and high reflectivity that characterize them. Secondly, non-emissive electronic displays refresh rates have to be further improved to the point of allowing fluid video content in large display devices. In the meantime, it expected that non-emissive electronic displays will thrive in areas of advertising and smart packaging, as well as in portable media applications. Nowadays, they already dominate the electronic book readers market. Even so, the potential of these displays is much greater. Their overall characteristics allow them to be incorporated in various types of surfaces, enabling the fabrication of dynamic environments (for example, in commercial spaces or health facilities), where visual information can be accessed in a seamless way.

It must be pointed out nonetheless that none of the display technology reviewed here meets all the technical requirements for every area of application. Hence, it is unlikely that a specific technology will completely dominate all the others in all possible market applications for displays. Instead, the dominance of certain technologies will be more evident in certain application whilst in others they will not thrive. Some technologies will simply be more suitable for certain applications than others.

Electrochromic displays present some distinctive features particularly relevant for the production of low cost, low power consumption display devices. The structural simplicity of electrochromic displays and the easiness which they are manufactured, together with the nature and availability of the raw materials used to produce this type of displays, makes them very well suitable for mass production as well as for Personal Fabrication. Indeed, the costs are expected to be low enough to render them practical for single-use applications. In addition, the displays are bistable and the average power consumptions are sufficiently low to enable their operation with small batteries. Also, electrochromic displays can be manufactured and assembled under normal environment conditions, not requiring special installations. It is expected that these characteristics will greatly contribute to the incorporation of electrochromic displays into all kind of objects, creating new products and new ways to experience and interact with digital information.

Nonetheless, electrochromic displays still need some improvements. For instance, it is necessary to overcome the long-term degradation of the electrochromic displays and thus, increase its lifetime. As Heikenfeld *et al.* (2011) point out, there is still a sizable gap between the theoretical performance of electrochromic displays and the actual capabilities of working displays.

4 System Architecture: Assembling Printed Electronics Visual Information Applications

4.1 Hardware Structure

One of the main concerns when developing the hardware structure for the various devices was to keep the hardware components to a minimum necessary. Several hardware structures were explored depending on the desired visual content and are here systematised. The assembly of the devices was done following an approach that intended to combine the principles behind Printed Electronics and Personal Fabrication. In the following subsections is explained in detail the assembly process, namely of direct addressing, passive-matrix and active-matrix electrochromic displays as well as of the required control units.

4.1.1 Display assembly

Electrochromic displays, as mention in section 3.2.1, are typically composed of five superimposed layers positioned between two protective substrates in a laminate configuration. The electrochromic layer and the electroactive layer are physically separated from each other by a solid, semi-solid or liquid electrolyte, and stacked between two opposing electrodes. The devices produced in the context of this thesis were assembled according to the following configuration, in normal ambient conditions (Figure 4-1):

$$\langle\,(\,PET \mid ITO) \mid PEDOT{:}PSS \mid\mid Electrolyte \mid\mid PEDOT{:}PSS \mid (\,ITO \mid PET\,)\,\rangle$$

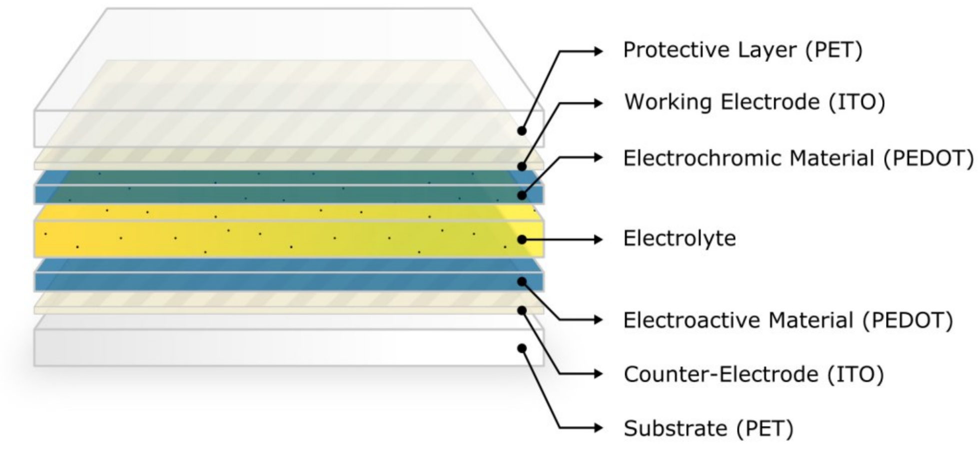


Figure 4-1: Assembly configuration of an electrochromic displays.

A commercially available transparent PET film, coated on one of the sides with ITO, was used both as the protective layer and substrate (PET layer) as well as the working electrode and counter-electrode (ITO layer). The electrochromic material (PEDOT:PSS) was deposited onto the ITO layer by means of a commercial inkjet printer, a Fujifilm Dimatix DMP-2831. Once both electrochromic and electroactive layers were printed, a 0,5 cm wide copper conducting tape was stick to the bottom edge of each functional layer $((PET\,|\,ITO)\,|\,PEDOT\!:\!PSS\,|)$ for electrical connection. A double-sided adhesive tape sheet, 1 mm thickness, was used to bond both layers as well as spacing element. It was cut like a frame and placed on the electrochromic layer so that it covered the four edges. The frame was filled with the electrolyte, and the second functional layer was mounted onto the first one in such a way that the two ITO coatings faced each other inside the assembled display. The surplus of electrolyte was squeezed out, wiped, and the edges firmly sealed. Finally, the sealed display was UV-cured for 10 seconds in order to solidify the electrolyte (by eliminating all the solvents and reaching complete polymerization). All the work was developed at Ynvisible facilities, in collaboration with their personnel. In the next section it is described in detail the function of each of the layers that composed the display.

4.1.1.1 Display components

4.1.1.1.1 Electrochromic and electroactive layers

Electrochromic devices can be regarded as a rechargeable electrochemical cell, where half-cell is in part composed by the electrochromic layer and the other half by the electroactive layer. When an electric field is applied to the electrochromic layer, it becomes absorptive to the electromagnetic radiation of a certain wavelength in the visible region. As a result, it becomes coloured. In turn, when the application of the electric field ceases, the electrochromic material changes back to its initial redox state along with its optical absorptions. Typically, this change is to a non-absorbent state in the visible spectrum, resulting in the electrochromic layer becoming transparent. In the case that both electrochromic states are absorptive in the visible spectrum, the electrochromic layer switches to another distinct colour. The complete oxidation or reduction of the electrochromic layer results in the greatest amount of optical contrast possible.

The main purpose of the electroactive layer is to counter balance the charges of the electrochromic layer upon the redox reaction. Nonetheless, the electroactive layer can also be electrochromic. In these cases, the colouration of the electroactive layer is induced when charging the electrochromic layer with an opposite charge of that of the electroactive layer. Hence, when an electrochromic reaction such as an electrochemical oxidation occurs at the electrochromic layer, a reduction reaction involving the same amount of charge transfer as in the oxidation reaction occurs at the electroactive layer (the electron transference is done in the reverse sense of that of the electrochromic layer).

The electrochromic material used as the electroactive layer can be the same as the one of the persistent electrochromic layer or different. The use of the same electrochromic material in both layers facilitates the assembly process, limiting however the number of possible colours. The superimposition of the two layers can be used to create, for instance, two complementary images that, depending on the electrochromic materials chosen, can have distinct colours. To successfully observe the colours transitions, both layers need to have a colourless transparent electrochromic mode.

Several chemical materials, both inorganic and organic, are known to show electrochromic behaviour and have been extensively investigated and reviewed on various publications covering the diverse categories of electrochromic materials and their applications (Granqvist, 1995; Monk et al., 1995, 2007; Mortimer, 1997, 1999, 2011; Mortimer et al., 2006; Rowley and Mortimer, 2002; Somani and Radhakrishnan, 2003). Whilst exist many types of chemical species that exhibit electrochromism, only those with favourable electrochromic performance parameters (see (Monk et al., 1995, 2007)) are potentially useful in commercial applications. Hence, most applications require electrochromic materials with high contrast ratio, high life cycle, high write-erase efficiency, low power consumption, high colouration efficiency and a response time adequate to the application (for example, electrochromic displays require fast response times in the order of few hundred milliseconds, whereas smart windows can operate with longer response times, in the order of several seconds). Another criterion than can be taken into account is the toxicity of the electrochromic material. Table 4-1 provides a list of the electrochromic materials commonly used in the fabrication of electrochromic devices. For more information, see Annex A.

Table 4-1: Commonly used electrochromic materials.

Transition Metal Oxides	e.g. Tungsten Trioxide: WO_3 e.g. Molybdenum Trioxide: MoO_3 e.g. Vanadium Pentoxide: V_2O_5 e.g. Niobium Pentoxide: Nb_2O_5 e.g. Iridium Hydroxide: $Ir(OH)_3$ e.g. Nickel(II) Hydroxide: $Ni(OH)_2$
Prussian blue systems	Prussian blue: $[Fe^{III}Fe^{II}(CN)_6]^-$ Prussian white: $[Fe^{II}Fe^{II}(CN)_6]^{2-}$ Prussian green: $Fe_3^{III}[Fe^{III}(CN)_6]_2[Fe^{II}(CN)_6]^-$ Prussian yellow: $[Fe^{III}Fe^{III}(CN)_6]$
Viologens	1,1'-di-substituent-4,4'-bipyridilium salts e.g. methyl viologen: 1,1'-di-methyl-4,4'-bipyridilium
Conjugated Conducting Polymers	Polythiophene and its derivatives: e.g. Polythiophene (PT) e.g. Poly(3,4-ethylene dioxythiophene) (PEDOT) e.g. Poly(3-methyl thiophene) (PMT) e.g. Poly(3-hexyl thiophene) (PHT) e.g. Poly(3-alkyl thiophene) (PAT) e.g. Poly(3,4-propylenedioxythiophene) (PProDOT) Polyaniline (PANI) Polypyrrole (PPy)
Metallopolymers	$[M^{II}(2{,}2' - bipyridine)_3]^{2+}$ compounds

The electrochromic displays produced in the context of this thesis all used PEDOT:PSS as the electrochromic material[20]. The thin films were prepared from a commercially available solution of PEDOT:PSS from Aldrich Chemistry (reference 483095), kindly supplied by Ynvisible. The deposition of the PEDOT:PSS film was done by means of a commercial inkjet printer (Fujifilm Dimatix DMP-2831)[21], at Ynvisible facilities. The choice of PEDOT:PSS as the electrochromic material was mainly because of its properties, in particular, its high chemical stability in normal conditions; its high optical contrast between the bleached and coloured state; and its low electronic bandgap; as well as due to the fact that it can be processed from a water emulsion and easily deposited in various substrate, rigid and flexible, by means of inkjet printing.

4.1.1.1.2 Ion-conducting layer: electrolyte

The electrolyte physically separates the electrochromic layer from the electroactive layer and thus, the working electrode from the counter-electrode. However, beyond functioning merely as a physical separator, the electrolyte layer is primarily an ion-storage layer, acting as a source and sink of cations and anions as the various redox processes take place. It is through the electrolyte layer that charged species are exchanged between the electrochromic and the electroactive layers during the redox process, and that the same process is balanced. As electrons are injected in one of the electrodes, the equivalent amount of electrons is extracted from the other. Eventually, the continuous flow of electrons results in the electrochromic cell becoming polarised. The ions present in the electrolyte layer counteract the polarisation by electrically neutralising the surplus of charges in the electrodes. To avoid short circuits in the electrochromic cell, both electrodes must be physically and electrically separated. Hence, the electrolyte layer must be not only ionically conductive but also an electrical insulator. The electrolyte layer has to be as well highly transparent and electrochemically inert over the voltage potential range needed to induce the colour transition, i.e. it cannot participate in the electrochemical reactions.

The electrolyte typically consists of a salt dissolved in a polymer matrix (see for instance, (Gray, 1991; MacCallum and Vincent, 1989)). The salt is responsible for the introduction of positive or negative charge carriers, and the polymer matrix serves as the medium through which they move between the polarised electrodes.

[20] See Box A-1 in Annex A for more information about PEDOT.
[21] See (Fujifilm Dimatix, 2008) for the printer datasheet.

The viscosity of such polymers increases with molecular weight, hence polymers range from liquid at low molecular weight to longer polymers that behave as rigid solids (Byker, 2001; Mortimer, 2011). As such, several types of electrolytes have been developed and used in electrochromic devices including liquid (Huang and Ho, 2006; Randin, 1978; Rocco et al., 1996; Schmitt and Aegerter, 2001), gel (Inaba et al., 1995; Kobayashi et al., 2007; Su et al., 1998; Vondrák et al., 1999), solid (De Paoli et al., 1997; Duek et al., 1993; Jee et al., 2011; Varshney et al., 2003), hybrid (Liang and Kuo, 2004; Orel et al., 2003; Rodrigues et al., 2011; Souza et al., 2007; Zelazowska et al., 2007) and ionic liquid (Brazier et al., 2007; Desai et al., 2011; Lu et al., 2002, 2003; Marcilla et al., 2006; Vidinha et al., 2008) electrolytes. The selection of an appropriate electrolyte system is fundamental in terms of operation and long term stability of the electrochromic device. Frequent stability problems associated with electrolytes include solvent volatility and electrolyte leakage. These typically arise from mechanical seal failures and ultimately result in the device failure. For more information on the various types of electrolytes, see Annex B.

4.1.1.1.3 *Working electrode and counter-electrode*

The working electrode and the counter-electrode are typically formed by an optically transparent, electrically conducting transition-metal oxide thin film. Indium tin oxide (tin-doped indium oxide or simply, ITO) (see (Ishiguro et al., 1958)) is currently the industry standard. The low electrical resistance and the high transmittance in the visible region of the electromagnetic spectrum, as well as the chemical resistance and the ease with which can be deposited as a thin film make ITO the transparent conducting film of choice, being widely used as electrode in most known flat panel display technologies, namely in liquid crystal displays, plasma displays, light-emitting diode (LED) displays, and electronic ink based displays, apart from electrochromic displays. As with all transparent conducting films, a compromise must be made between the conductivity and the transparency of the thin film. By decreasing the thickness of the film it is possible to improve its transparency, however, this will consequently result in a decrease of the concentration of charge carriers and thus in a reduction of the film conductivity. ITO thin films are commonly deposited on surfaces by electron beam evaporation, physical vapour deposition, or a range of sputter deposition techniques.

Despite the attractive properties, the use of ITO as the choice material in these applications may be limited for both technical and economic reasons. The limited supply of indium, and its consequent high cost, along with the ever-increasing number of devices requiring flat panel displays, started to contest the viability of using such a rare element for transparent conducting films. Furthermore, ITO is a brittle, crystalline material that when stressed can fracture, irreversibly dropping the film conductivity by several orders of magnitude and thus limiting its utility in applications that require some flexibility. Hence, alternatives are in high demand (Demming, 2012; Wassei and Kaner, 2010). Novel alternatives include the use of organic films developed using carbon nanotubes (Nakayama and Akita, 2001; Wu et al., 2004; Zhang et al., 2006) or graphene (Lahiri et al., 2011; Wang et al., 2010; Wassei and Kaner, 2010), as well as conductive polymers such as PEDOT and its derivatives (Y-B Kim et al., 2009; Wang and MacDiarmid, 2007), or thin foils of silver (De et al., 2009; Zeng et al., 2010) or copper (Kang et al., 2010; Rathmell et al., 2010) nanowires.

The optically transparent electrodes are typically coated on a transparent substrate such as glass or plastic, although other materials like paper, wood, ceramic, or cork, with different optical properties, can also be used. It must be noted, nonetheless, that at least one of the electrodes has to be transparent.

4.1.1.1.4 Protective layer and substrate

Practical applications of electrochromic devices require that these be able to resist the adverse conditions of the environment in addition to withstand everyday physical use. The exposure to atmospheric oxygen and water, as well as to other environmental contaminates such as ammonia, chlorine, or hydrogen sulphide, can degrade the performance of electrochromic devices and reduce their lifetime. Hence, electrochromic devices must be built not only to prevent internal materials from escaping the device, but also to prevent external materials from entering. The protective layer and the substrate provide the required barrier against the deleterious effects of ambient oxidants as well as protection against mechanical damage, in addition to function as a support structure for the different components of the electrochromic device. The sealant used around the electrochromic device must also withstand the adverse environmental conditions and be chemically stable.

Traditionally, the protective layer and substrate consist of a laminate glass pane which can be heat-strengthened or tempered, or of a plastic film, such as polyethylene terephthalate (PET). Glass has been widely used mainly because of its better thermal stability, though devices built on glass are rigid. For flexible display, plastic films are the choice. The first report of an electrochromic device using plastic substrates was made by Antinucci and co-workers in 1995 (Antinucci et al., 1995).

A preferred embodiment would employ the protective layer transparent, whereas the substrate, depending on whether the displays functions in transmissive or reflective mode must be, or not, transparent. As already mention in the previous section, other materials like paper, wood, ceramic, or cork, with different optical properties, can also be used if prepared adequately to have a conductive surface.

4.1.2 Drive circuit and control unit

The drive circuit represents here the electrical circuit required to control the colouring and erasing process of each picture element of an electrochromic display. Typically, it comprises:

- a circuit for applying a constant voltage potential selectively to each picture element working electrode, for colouring purposes;

- a second circuit for short-circuiting the electrodes of a coloured picture element, for erasing purposes (i.e. for reversing the state of the picture element);

- a third circuit for keeping the device at open-circuit conditions, for enabling the memory effect of the electrochromic cell.

The control unit, normally a microcontroller, is responsible for generating the control signals for activating the picture elements. Ideally, the system should have a potential control means and a power blocking means to prevent cells from being overdriven. If a coloured cell is exposed to a high voltage sequence for a long period of time, it can be destroyed. Hence, during operation, each picture element should be driven cyclically for a predetermined short period of time and in between cycles maintained in its open state, taking advantage of the memory effect.

The simplest drive circuit possible is a two-terminal drive circuit connected to a 1.5V battery, with a reversing switch (Figure 4-2). In this setting, the operation of the circuit is manual, although it can be automated electronically with a capacitor or a simple microcontroller (e.g. a 555 timer integrated circuit). It must be pointed out, nonetheless, that this type of drive circuit is essentially adequate for simple, one cell, displays. Displays with complex architectures will obligatorily require an adequate control unit to manage the state of the various picture elements that compose the display.

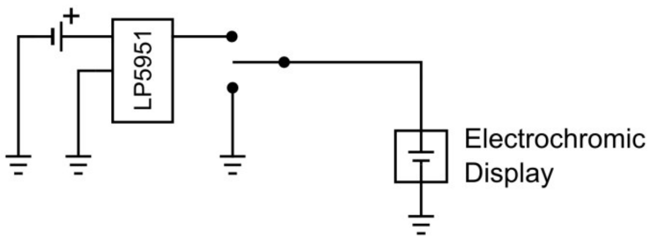


Figure 4-2: Schematic representation of a drive circuit for a simple electrochromic display controlled by a toggle button.

The initial electrochromic display prototypes developed by the author when exploring the various addressing schemes possible (see next chapter) had as a control unit an adapted Arduino Mega board[22]. The main rationale behind this approach was to use an open-source platform that could be easily purchasable and easily programmable by anyone.

4.1.2.1 Addressing methods

Addressing can be defined as the process by which individual picture elements in a display (generally pixels) are switched on or off in order to create an image on the display. There are three main different addressing methods by which this can be done: direct addressing, passive-matrix addressing and active-matrix addressing (Cristaldi et al., 2009; Kelly and O'Neill, 2000). The implementation of a particular addressing method in an electrochromic display is closely related to its desired application, the information content to be displayed and the production cost. The display area and the response time can also have influence in the choice, however, generally are not decisive. The selection of one method over other influences the architecture and construction of the display, as the operation of the microcontroller responsible for controlling the display.

[22] Arduino is an open-source electronics prototyping platform based on easy-to-use hardware and software. The hardware consists of a small microcontroller board, commonly known as Arduino board, set up with an Atmel AVR processor and on-board input/output (I/O) support. The software consists of a standard programming language compiler known as the Arduino IDE (Integrated Development Environment), used to program the Atmel AVR processor. The Arduino boards can be easily purchased online or in any electronics store and they are used by hobbyists and makers everywhere in the word. For more information on the Arduino open-source platform, see (Banzi, 2011; Margolis, 2011).

4.1.2.1.1 Direct addressing

In direct addressing displays, each single controllable picture element, commonly named segment, has its own drive circuit. As such, each segment is directly connected to the display microcontroller and, in order to switch it on or off, the microprocessor separately applies a control signal to that specific segment. The overall image displayed is produced based on the spatial arrangement of the active segments, which can be based on simple geometric figures or custom pictures, such as, different pictograms.

The most well known application of direct addressing is the seven-segment arrangement (Figure 4-3) used in alphanumerical displays such as the ones present in simple calculators and digital watches. In this type of arrangement, the segments normally have a uniform shape and size, being frequently elongated hexagons, though rectangles and trapezoids can as well be used. Their spatial arrangement is done in such a way that through the combined activation of the different segments it is possible to reproduce the Arabic numerals. Several seven-segment arrangements can be combined together to form a composite display to show more digits. However, the increase in the number of segments means a higher number of drive circuits.

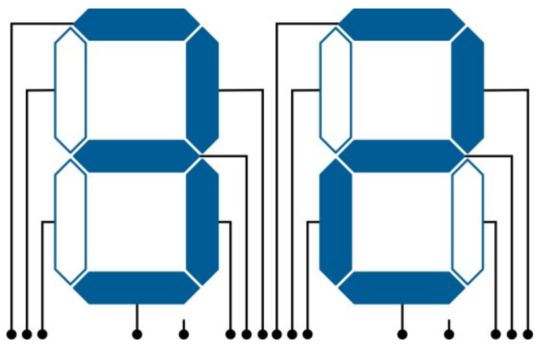


Figure 4-3: Illustration of a two-digit seven-segment display configuration

The implementation of seven-segments arrangements in electrochromic displays have been reported in literature since the early seventies (Itaya et al., 1982; Sampsell, 1981; Schoot et al., 1973; Shay et al., 1982) and are disclosed in several patents (Duchene, 1982; Hamada et al., 1979; Huguenin, 1981; Iwata, 1985; Shimizu and Fujikawa, 1979). Various examples can be found using distinct electrochromic materials as well as diverse carrier substrates and fabrication techniques. Still, only recently the first commercial devices started to appear.

For the purpose of this thesis, several direct addressing electrochromic displays were constructed and tested. The overall assembly procedure was done as described in section 4.1.1 with the variation that the electrical paths required to control each segment had to be patterned, in advance, directly into the display ITO electrodes layers. The process consists of sectioning the ITO film, in a number of sections equal to the number of segments in the display. Each section encloses one segment and forms a path electrically isolated from the others which is connected directly to the display peripheral microcontroller. The sectioning was done either by using a manual scalpel or by means of a laser cutter (Epilog Mini 24 - Legend Elite Series)[23]. In Figure 4-4, which schematically exemplifies the assembly of the seven-segment electrochromic display, the circuit paths correspond to the delimitation of the dashed lines.

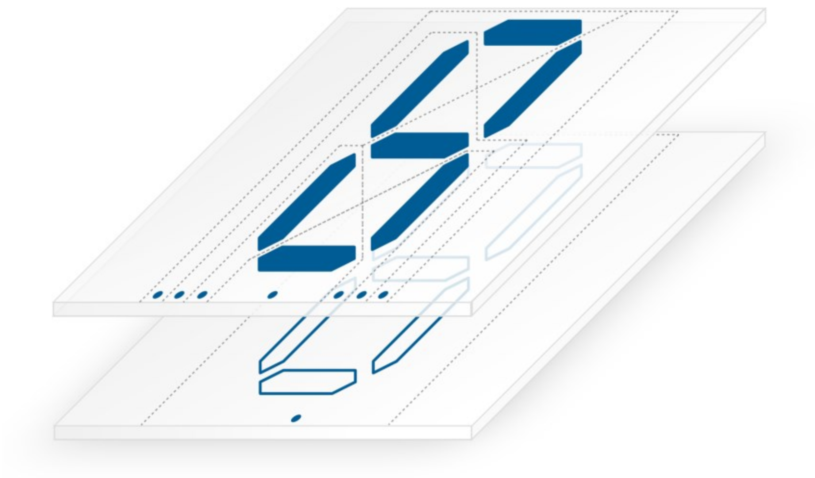


Figure 4-4: Schematic representation of the configuration of a seven-segment electrochromic display.

Each segment is formed by two adjacent films of PEDOT:PSS, deposited over a transparent ITO electrode: one placed over the substrate (bottom electrode), and the other placed beneath the protective layer (top electrode). The bottom electrode is common to all the segments, forming a single circuit path with a single connection point (the display reference point or ground). The top electrode is divided according to each segment, with each path having its own connection point (blue dots on the top layer, Figure 4-4). Between the two electrodes, as in any

[23] See (Epilog Laser, 2009) for the technical specifications, and Annex C for the cutting parameters.

typical electrochromic cell, is an ionically conductive electrolyte that is common to all the segments (not represented in Figure 4-4 for simplification purposes). The intersection of the top electrode with the bottom electrode results in a display segment. Alternatively, it is possible to follow an approach in which the ITO film is replaced by a non-conductive layer and a circuit line connecting each segment to the microcontroller is directly printed in the non-conductive layer using conductive ink. The advantage of the first approach (i.e. the one followed) is in the display being fully transparent since the connection paths are not visible. On the other hand, the response times of the displays were slightly slower (approximately 1s).

A crucial aspect of the development of the control unit for the electrochromic displays was that it had to be entirely end-user-programmable. As such, an Arduino Mega board with a built-in Atmel ATmega1280 microcontroller was adapted to be used as the display control unit. The integration of the drive circuit (Figure 4-5) into the Arduino Mega board was done by means of a custom made shield board that sit atop the Arduino Mega board.

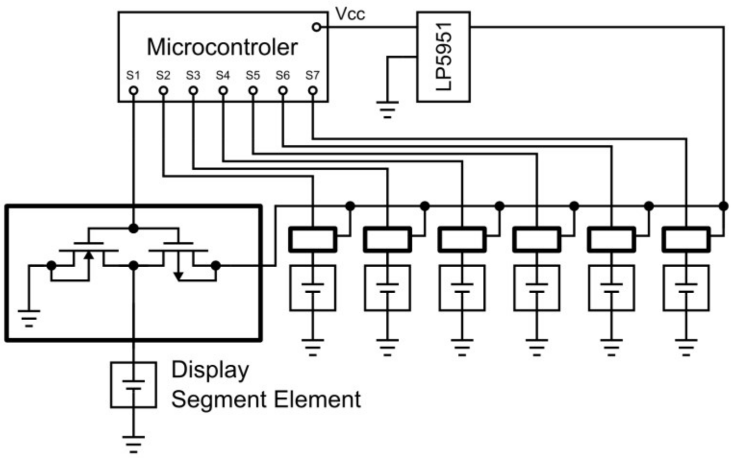


Figure 4-5: Schematic representation of a seven-segment, direct addressing electrochromic display drive circuit.

The drive circuit, which was developed in collaboration with Ynvisible, consisted essentially of a voltage regulator (LP5951), which was responsible for converting the microcontroller 5V operating voltage into the 1.5V required to operate the display segments, and seven sets of two field-effect transistors (one per each segment), which were responsible for switching the electronic signals. Each set of field-effect transistors was connected to a digital input/output (I/O) pin of the Arduino board. Addressed segments were keep at a negative potential of 1.5V, while non-addressed were put at a potential of 0V. The voltage of all the segments was updated simultaneously.

For testing and demonstration purposes, the Arduino board was connected to a desktop computer via serial interface. The necessary software for communicating with the microcontroller was programmed specifically for this purpose in the Processing programming language (see Annex D). It allowed users to activate individually each of the display segments as they seemed fit, or alternatively, to choose a number and see it reproduced on the electrochromic display. Regardless, the drive system was as well capable to operate fully independent from the control of the desktop computer.

Figure 4-6 illustrates one of the direct addressing electrochromic displays prototypes produced: a typical seven-segment display capable of reproduce the numbers from zero to nine (as shown in Figure 4-23) exhibiting a representation of the number three.

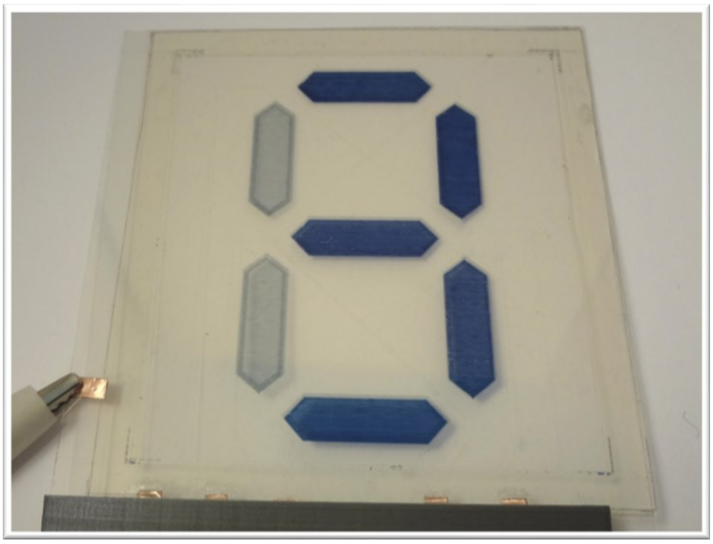


Figure 4-6: Examples of direct addressing electrochromic displays.

For an improved energy efficiency, the display should take advantage of the memory effect and be refreshed in cycles, opposite of being constantly on. Hence, as soon as the driven segments are coloured, the writing action should automatically stop and the image would prevail without any current input. After a pre-determined period of time, when the segments colour density weakens, the writing action starts again, recommencing the cycle. Also, if a coloured segment is exposed for too long to a high voltage, or alternatively to an excessive voltage, the segment will be overdriven and, eventually, visually damaged.

Display format, response time, and contrast are all features that can vary depending on the desired use. Factors such as the size of the segments, and for that matter of the display, the ohmic resistance between segments and connection points, as well as the applied voltage strongly influence the response time. Higher colour contrasts and image contrasts can be achieved by printing additional layers of electrochromic material or by using a versatile driver that allows the control of the signal amplitude individually for each pixel. Because colouration in electrochromic devices is an electrochromic reaction, colour density is proportional to the quantity of electricity flowing in the display electrode per square area. If too many segments are driven simultaneously at an equal voltage for an equal period of time, each segment can have a different colour density depending on the size of the segment or on the number of segments that are driven simultaneously. Possible solutions include adjusting the output impedance of each driving circuit in proportion to the number of simultaneously driven segments, as well as in accordance to the segments areas, or using different driving times for each segment.

Overall, direct addressing is essentially convenient for display applications where there are only a reduced number of elements that have to be activated. A display with n segments requires a total of n drivers with $n+1$ connections (one for each segment, in addition to one for the reference point). Increasing the number of segments to further increase the information content of the display can significantly raise the number of connections to a point where managing all becomes too complicated. For example, if we consider a typical seven-segment display, to replicate a simple digital clock with six digits, using a direct addressing approach, a minimum number of 42+1 connections are required. However, if the display must be able as well to display text, a fourteen-segment scheme must be implemented (at least), raising the number of connections to 84+1. It is possible to partially diminish this problem by using a multiplexed approach, where the anodes of equivalent segments of different digits are connected together and to a single driver, while the cathodes of all segments for each digit would be connected separately. In this way, a six digit display with seven segments would require only 6 cathode drivers and 6 anode drivers. This approach requires, nonetheless, that each single digit be operated sequentially: to activate a certain number of segments on a particular digit, the cathode driver for the selected digit and the anode drivers for the desired segments have to be enabled. In order then to activate the segments of a second digit, the cathode driver of the first one has to be disabled, so that the second digit can be enabled, and the new segments activated.

The use of direct addressing in the control of dot-matrix displays, in principle capable of higher resolutions, further emphasises the scaling limitations of direct addressing. A matrix display with $m \times n$ elements requires $m \times n+1$ connections. While a small matrix, like the one represented in Figure 4-7, is still feasible, as the number of elements of the matrix increases, the display becomes unpractical due to the high cost of using so many drivers and the absence of space between elements for the higher number of connections. Consequently, direct addressing displays are mainly used in low information content applications. High information content and graphical applications require higher resolutions which simply are not feasible through direct addressing.

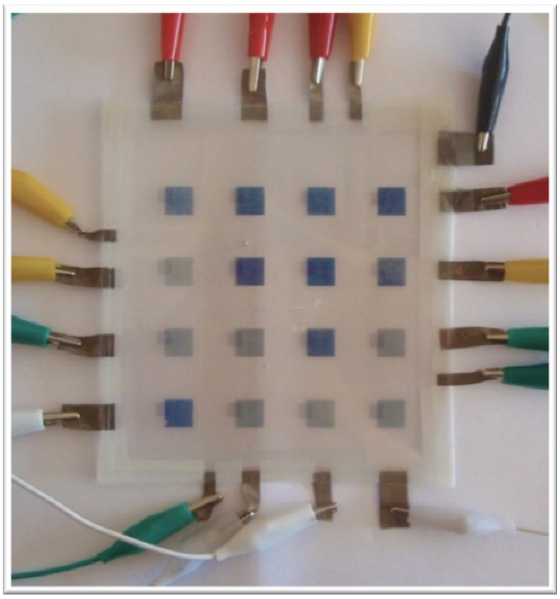


Figure 4-7: Direct addressing 4x4 electrochromic matrix display.

4.1.2.1.2 <u>Passive-matrix addressing</u>

The passive-matrix addressing scheme appears as a solution to the limitations of direct addressing, in particular to the need to present high information content in displays (Cristaldi et al., 2009; Kawamoto, 2002). This was mainly achieved by moving to a scheme composed by pixels arranged in a matrix with row and column electrodes (see Figure 4-8).

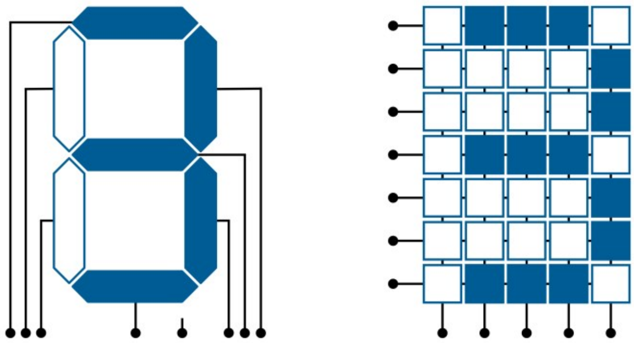


Figure 4-8: Comparison between the layout of a seven-segment, direct addressing arrangement and the layout of a passive-matrix arrangement.

The novelty of the scheme was in the pixels being addressed by their row and column instead of being driven separately (Figure 4-9). Hence, in a passive-matrix display with *m* rows and *n* columns, only *n+m* connections are required for activating the *mxn* pixels in opposition to the *m×n+1* connections required using the direct addressing approach. The method was initially developed for Liquid Crystal Displays, though, it was easily adapted to other types of displays, namely electrochromic displays.

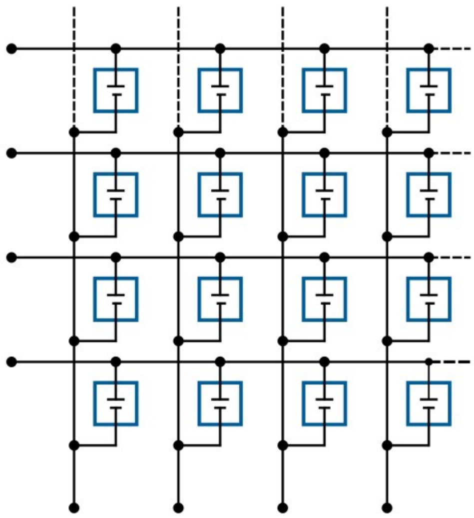


Figure 4-9: Circuit diagram for a passive-matrix display.

To activate a specific pixel in a passive-matrix display, a carefully timed control signal is sent across the row containing the selected pixel, while the corresponding column is connected to ground. By applying the control signals sequentially, row by row, each individual pixel within the entire display matrix can be uniquely addressed. An external microcontroller is responsible for controlling where and when the control signals are applied. For the method to function, each pixel in a passive-matrix display has to be capable of maintain its state without active circuitry until it can be refreshed again, i.e. it has to be bi-stable. If the refresh time of the entire display is shorter than the turn-off time, multiple pixel images can be formed by activating the appropriate pixels. However, due to the fact that each row has to be addressed sequentially, the response time to turn the pixels on and off in an electrochromic display is usually too slow for full-motion video and can produce a ghosting effect when refreshing the full display.

Several passive-matrix displays prototypes were developed for the purpose of this thesis. Again, the overall assembly procedure was done as described in section 4.1.1 with the variation that the top ITO electrode was divided, either by using a manual scalpel or by means of a laser cutter (Epilog Mini 24 - Legend Elite Series), into columns and the bottom ITO electrode into rows (as illustrated in Figure 4-10 by the dashed lines). The intersection of a column with a row resulted in a pixel element, printed in PEDOT:PSS on each of the transparent ITO electrodes. Between the two electrodes was an ionically conductive electrolyte (not represented in Figure 4-10 for the purpose of simplification), which was sectioned according to each pixel with double-sided adhesive tape. The rows and columns were connected to the display drive system.

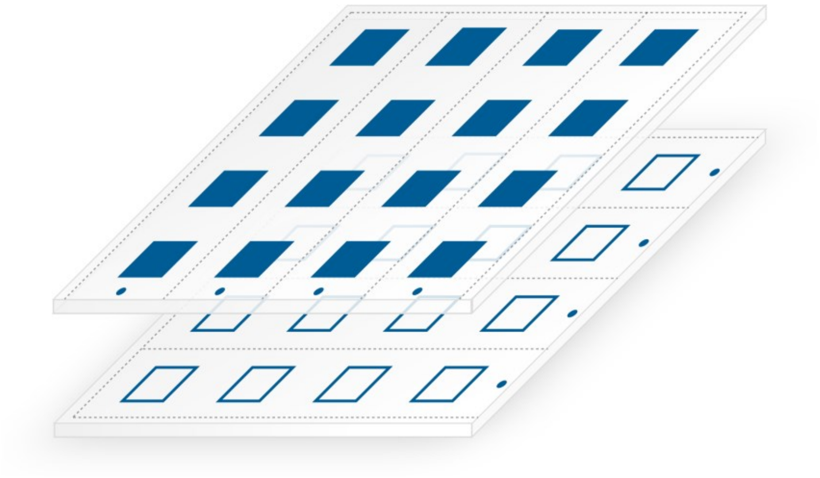


Figure 4-10: Schematic representation of the configuration of a passive-matrix electrochromic display.

The electrochromic display was controlled using a similar setup to the one implemented for the direct addressing scheme: an Arduino Mega board with a built-in Atmel ATmega1280 microcontroller served as the control unit, and the drive circuit (Figure 4-11) was integrated into the Arduino Mega board by means of a custom made shield board that sit atop the Arduino Mega board. A LP5951 voltage regulator was used, likewise, to convert the microcontroller 5V operating voltage into the 1.5V required to operate the display, whereas, eight sets of two field-effect transistors, one for each row and column, was responsible for switching the electronic signals. Each field-effect transistor, in opposite to each set, was connected to a digital I/O pin of the Arduino board. As a result, three different logic signals could be applied to each row and column: a low state (0V), a high state (±1.5V), and a high-impedance state (open circuit). The necessary software for communicating with the microcontroller was programmed specifically for this purpose in Processing (see Annex E).

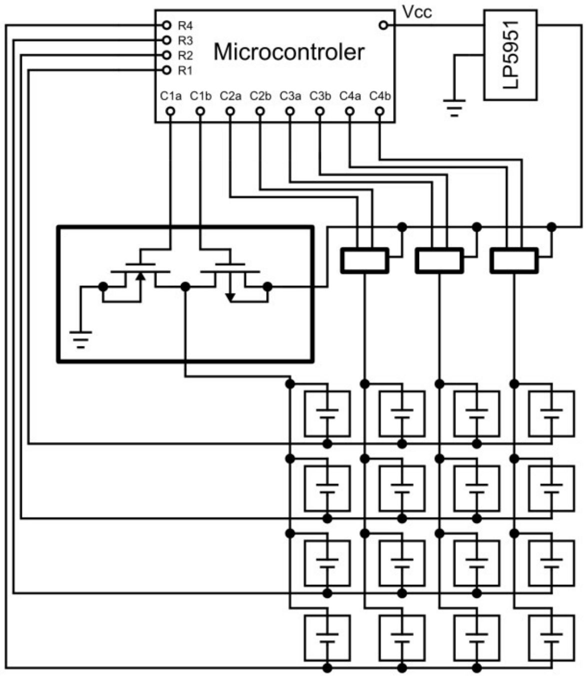


Figure 4-11: Schematic representation of a 4x4 passive-matrix electrochromic display drive circuit.

To activate a specific pixel, two different voltage signals are used on the rows and columns electrodes. The addressed row, i.e. the row where the pixel to be activated is located, is placed at a V_{set} potential (i.e. in the high state) while all others rows in the matrix are unselected with a V_{unset} potential (low state), whereas the addressed column is placed at a V_{on} potential (low state) while all the others columns are in a V_{off} potential (high state) (Figure 4-12). As only a single row is addressed at a certain time, the matrix is updated sequentially. The update cycle is finished when the whole matrix has been scanned through and all the rows have been addressed once. After that, in order to kept the pixels active, the same sequence of voltage signals can be repeated applied, or the matrix can be placed at the high-impedance state and take advantage of the memory effect of the electrochromic material. The latest offers improved energy efficiency.

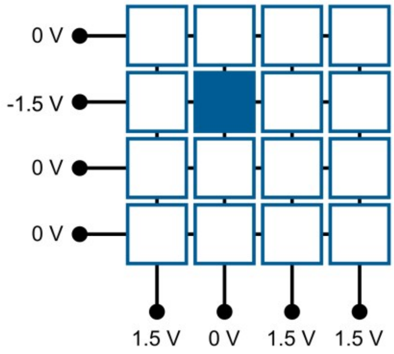


Figure 4-12: Pixel activation mechanism on a passive-matrix electrochromic display.

When multiple pixels in a row are simultaneously activated, it is possible to observe that active pixels get a slighter stronger colour in rows where the number of active pixels is higher. Adjusting the driving voltage of a row in accordance to the number of active pixels in that row can ensure the colour contrast uniformity by enhancing the colouration of the weakly coloured pixels (Edwards, 2005).

Figure 4-13 shows one of the passive-matrix electrochromic displays produced, a simple dot-matrix display with four rows and four columns.

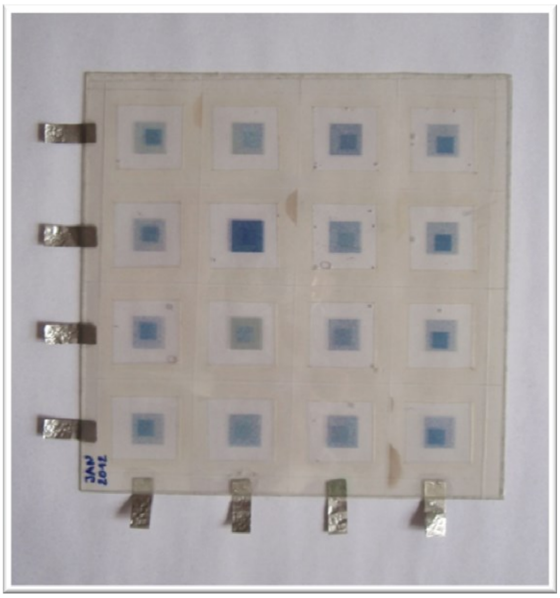


Figure 4-13: Example of a 4x4 passive-matrix electrochromic display.

The passive-matrix method enabled the construction of displays capable of presenting high information content with a minimum number of connections. Furthermore, it brought the advantage of a simplistic cost-efficient manufacturing process easily scaled to produce large dimension displays. However, it also presented some inherent issues that ended up limiting its practical applications. When the method was initially developed for Liquid Crystal Displays, it was noticed that as the size of the display increased, the image contrast ratio decreased and the response time slowed down (Kawamoto, 2002). Increasing the driving voltage to compensate the loss of contrast created a secondary problem: the colouration of undesired elements. When a pixel is being coloured, even though only one row and one column are being addressed, there are voltage leaks to neighbouring pixels due to the interaction between pixels in the same row and column electrodes. The increased drive voltage results in the selected pixel becoming coloured as desired, but also in the surrounding pixels becoming undesirably partially coloured. The effect is commonly known as crosstalk. The partially coloured pixels reduce the display contrast and diminish the image quality. Hence, a trade-off between resolution and contrast must be made.

The maximum number of rows (N_{max}) a passive-matrix display can have can be calculated using equation (4.1) (Alt and Pleshko, 1974), where V_{th} is the threshold voltage and ΔV is the transition voltage, i.e., the difference between the minimal voltage required to colour a pixel and the maximum voltage to erase it.

$$N_{max} = \left(\frac{V_{th}}{\Delta V}\right)^2 \qquad (4.1)$$

In electrochromic devices, the cross-talk effect can be minimised, first, by physically isolating each pixel against ionic current through the electrolyte layer, and second, by increasing the voltage threshold on the electrode interface (see (Aliev and Shin, 2002; Chang and Howard, 1975)).

Another possible solution to reduce the effect of cross-talk and increase the number of rows in a passive-matrix display is to adapt a “dual-scan” system: the column electrodes are divided into two sections, at midway point, and both upper and lower sections are refreshed separately. As a result, the maximum number of addressed rows is doubled, and the refresh rate is improved. In a similar approach, Bauwens *et al.* (2009) propose instead the use of a fully modular display, where each module acts as an individual passive-matrix display, having its own display driver. Each independent module would be connected to a central control unit that would be responsible for managing all the modules. According to the authors, this approach would remove the display size limitation, meaning the resolution and contrast can be made much higher. Also, as each module is refreshed in parallel, the overall refresh time of the display would also be greatly improved, i.e. reduced. Another advantage would come from the fact that, as each module is individually controllable, when displayed information changes, the display can be updated only in the affected modules.

Nowadays, passive-matrix addressing is essentially used in small sized displays such as the ones present in personal digital assistants (PDAs) and mobile phones.

4.1.2.1.3 Active-matrix addressing

The concept behind the active-matrix addressing scheme goes back to 1971 when Lechner, Marlowe and co-workers (Lechner et al., 1971; Marlowe and Nester, 1972) proposed the idea of using an array of thin-film transistors (TFTs) to control the pixels in Liquid Cristal Displays. The aim was to overcome the multiplexing limitation of the passive-matrix addressing scheme and eliminate the crosstalk effect (Kawamoto, 2002).

The construction of an active-matrix display is similar to the passive-matrix, with pixels arranged in a matrix with row and column electrodes. The key difference lies in the active-matrix display having a thin-film transistor built into each pixel, at the cross point of the row and column electrodes (see Figure 4-14). The TFT acts as a switch, precisely controlling the voltage of each pixel. To activate a specific pixel, first, a control signal is applied to the row where the pixel is located. This disables the TFTs located at that row and enables the flow of charge from the column electrode. Thus, by applying an appropriate voltage potential to a specific column, the respective pixel is activated. Once a row is updated, the transistors are changed back to their active state and the following row in the matrix can be addressed: the display is updated one line-at-a-time. The updating procedure is repeated until all the rows in the matrix have been addressed.

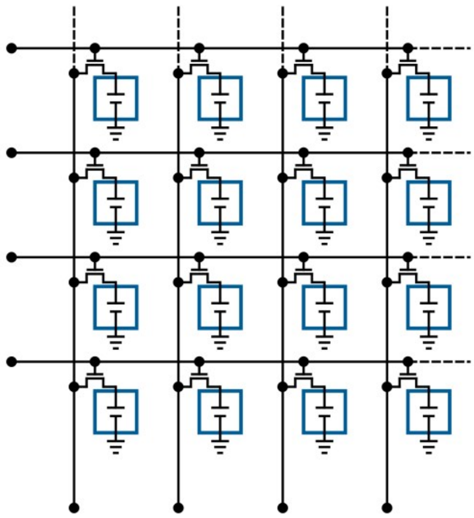


Figure 4-14: Circuit diagram for an active-matrix display.

Andersson *et al.* (Andersson et al., 2002, 2007), for example, developed an all-organic active-matrix addressed electrochromic display made on flexible substrates (Figure 4-15).

Figure 4-15: Example of a 5x5 active-matrix electrochromic display. Source: (Andersson et al., 2007).

Each individual pixel device was formed by combining an electrochromic display cell with an electrochemical transistor in a side by side arrangement (Figure 4-16). One of the interesting aspect of this display device was in the same organic and electrochemically active material, in this case PEDOT:PSS, being used as both the electrochromic display cell and the electrochemical transistor, as well as the conducting lines of the integrated active-matrix.

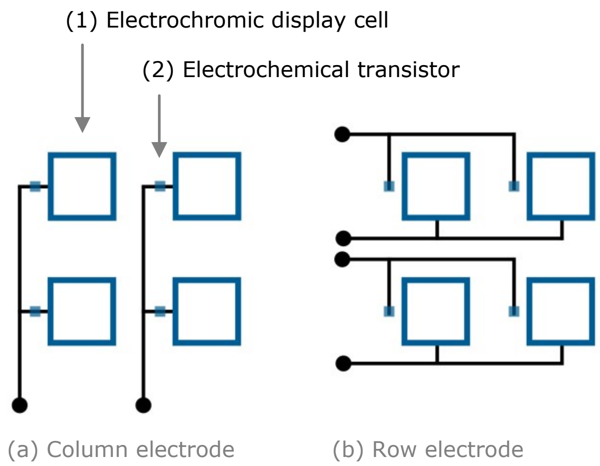


Figure 4-16: Schematic illustration of the column and row electrodes in a 2x2 active-matrix electrochromic display. Source: adapted from (Andersson et al., 2007).

The electrochemical transistor served as the switch to control the current flow to and from the electrochromic display cell. It consisted of a three-terminal transistor, where each terminal corresponded to one of the following electrodes: the source, the drain and the gate. A stripe of PEDOT:PSS served as the transistor channel, connecting the two associated electrodes, the source and the drain. The gate electrode was represented by an adjacent layer of PEDOT:PSS patterned close to the transistor channel. The gate electrode and the transistor channel were connected, ionically but not electronically, via a patterned electrolyte layer.

According to Andersson and co-workers, the described arrangement made it possible for an active-matrix display that is very cost effective and simple to manufacture (for example, using a roll-to-roll production procedure). Nonetheless, it also presents some drawbacks. Since the electrochemical transistor and the conducting lines are made from PEDOT:PSS, they change colour during the switching operation associated with the control of each pixel device. As a result, the display cannot be made transparent, and each pixel must be inside an opaque frame in order to conceal this issue. Moreover, the colouring area of each electrochromic display cell is affected by the area occupied by the electrochemical transistor.

Other examples of active-matrix electrochromic displays can be found in relevant literature (Chung et al., 2010; Tam et al., 2006) or referenced in patents (Cassidy, 2008; Fischer and Mathea, 2006; Green and Nicholson, 1991). These, however, usually involve using the traditional n-channel and p-channel metal-oxide-semiconductor TFTs and typically placing them at the display backplane. The biggest advantage in this approach, i.e. in placing the TFTs in the display backplane, is in the maximisation of the viewable pixel area.

In sum, the active-matrix addressing scheme solved the problems associated with passive-matrix displays, such as the scanning limitations and the crosstalk effect, as well as the contrast ratio and grey scale limitations. However, the cost and complexity of manufacturing an active-matrix display is also higher than a passive-matrix. Hence, its implementation should be pondered according to the type of application.

4.2 Types of content

The possible applications for electrochromic displays are numerous: from being integrated in traditional print magazines and posters to products packages and point of purchase (POP) locations or in signage systems. However, its successful deployment is greatly dependent on its visual content. It must be exciting and relevant, captivating and engaging users and consumers.

This section addresses the topic of content formats in electrochromic displays, focusing on the creation of interactive and dynamic visual content. Based on the different addressing methods described in section 4.1.2.1 and on the potential to create dynamic content and animations, six different formats of visual content were distinguished. Table 4-2 summarises the different categories of visual content and the necessary architectures for displaying it. The designation of each category intends to capture the uniqueness of that specific category in relation to the others.

In the following sub-sections, the six different types of content as well as the necessary architecture to reproduce them are described in detail. It provides a first glimpse of what could emerge as an inexpensive and creative platform for bridging the worlds of bits and atoms. The examples developed for each type of architecture/content illustrate how these can be implemented as simple, task-specific applications (though they are still rough prototypes) following an end-user fabrication process. All the materials and electronic components used to make the devices are easily purchasable online by anyone or in any electronics specialty store.

Table 4-2: Electrochromic displays visual content formats.

Type	Designation	Description	Addressing Method	Suitable for
I-A	Two-Status Still Image	Pre–determined visual content composed by a fixed image with two distinct statuses.	Direct	Entertainment Information
I-B	KeyFrame Animated Image	Pre-determined visual content composed by an animation sequence based on a fixed image.	Direct	Entertainment Information
II-A	Plain Segment-based Dynamic Image	Dynamic visual content produced by individually activating various abstract segments in order to form an image.	Direct	Information
II-B	Multi Pictograph-based Dynamic Image	Dynamic visual content produced by individually activating various pre-determined pictographic segments.	Direct Matrix	Entertainment Information
III-A	Pixel-based Dynamic Image	Dynamic visual content composed by individually addressable abstract pixels.	Matrix	Entertainment Information Simulation
III-B	Multi-Concept Pixel-based Dynamic Image	Dynamic visual content composed by individually addressable sets of pixels representing specific concepts.	Matrix	Entertainment Information Simulation

4.2.1 Type I-A: Two-status still image

Type I-A represents the simplest possible architecture for presenting visual information in an electrochromic display. It consists of a fixed, still image object (e.g. a picture or a pictogram) that can either be erased (i.e. made invisible) or replaced by another pre-fixed image. The use of a secondary overlapping image that complements the first one in a way that it adds some feature or alters some of its elements can allow the creation of simple pre-determined animations. For example, a very simple illusion of movement can be created to some extent by switching between the two images in a rapid succession. Figure 4-17 illustrates this concept. The effect achieved recalls the animations obtained in early animation devices of the 19th century such as the thaumatrope or the zoetrope[24], in which the basic animation repeats itself in an endless loop, beginning over and over again. Other sort of effects and transitions can be implemented, for instance, to highlight and draw attention to important information, events or products, or simply to function as decorative visuals.

[24] See (Furniss, 2008) for an introduction on early motion devices.

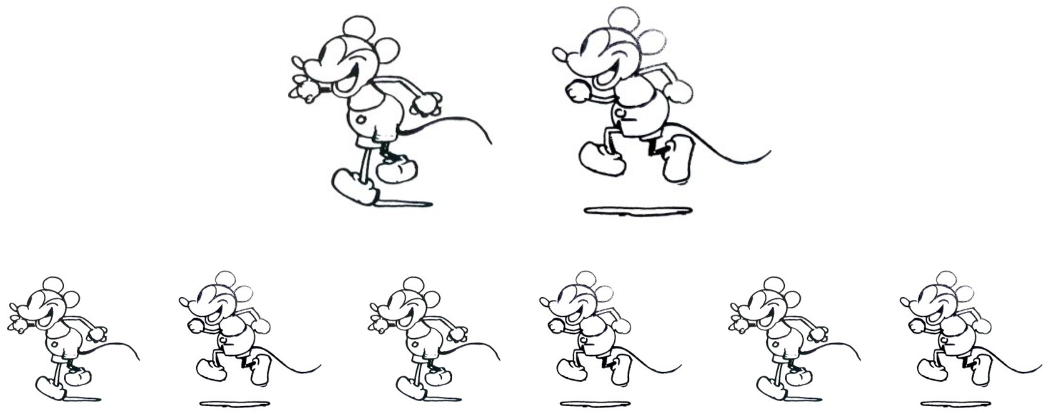


Figure 4-17: Illustration of a simple two-frame animation (animation flows from right to left). Source: adapted from (Thomas and Johnston, 1997).

The architecture of type I-A displays is based on one single electrochromic cell in which the first image is printed on the electrochromic layer whilst the second image is printed on the electroactive layer. A two-terminal drive circuit connected to a 1.5V battery, with a reversing switch (as described in section 4.1.2) can be used to control the display.

Figure 4-18 illustrates a type I-A electrochromic display embedded in a postcard. When a button on the postcard is pressed, the electrochromic display is activated and the hidden image becomes visible, showing its content, in this case, for a limited period of time.

Figure 4-18: Example of a type I-A electrochromic display - Electrochromic postcard. Source: (Ynvisible, 2012).

The limitations in terms of dynamic content inherent to this type of displays naturally make its use only viable in certain pre-determined contexts. As the visual content cannot be altered, it must be specifically defined beforehand based on the desired application. For example, a real-life application could be the implementation of this type of displays as warning signs, being only activated to alert bystanders when specific situations or problems arise.

4.2.2 Type I-B: Keyframe animated image

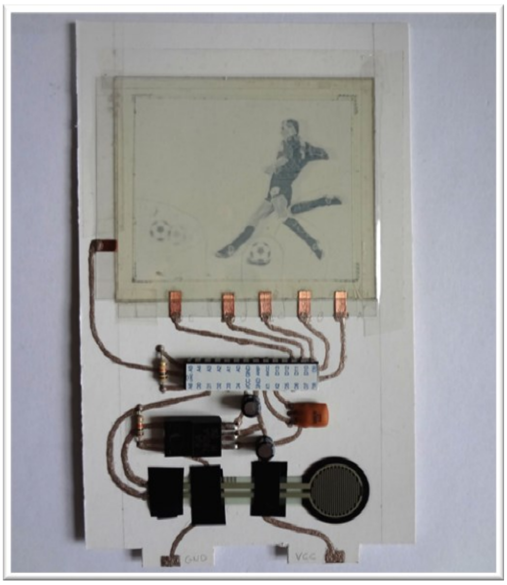
The distinctive feature of type I-B electrochromic displays is that they are structured to allow the creation of motion from a preset image sorted in various key sequences. The pictorial object has dynamic motion but is pre-determined. Each key sequence, or keyframe, can be activated independently and represents a specific instance that occurs at a certain point in the pre-determined animation. By programming sequences of switching keyframe on and off, it is possible to form dynamic patterns that form animated images. When the keyframes are activated in the correct sequence, the overall animation is visually reproduced.

Figure 4-19 illustrates a pre-determined sequential animation structured in three keyframes. A still image is used as a common element in the three distinct complementary keyframes which, when activated in the correct order originate a continuing animated action. In this specific case, they create a simple animation of a football player kicking a football ball. The animation is composed by a sequence of movements with a beginning and ending though it is possible to devise cyclic animations that repeat over and over.

Figure 4-19: Example of a type I-B electrochromic display – Football player.

Figure 4-20A depicts the final display while Figure 4-20B shows the electronic circuit assembled to operate it built-in a cardboard base frame. A copper-base conductive ink was used to trace the circuit lines. When the button coloured blue in the picture is pressed, the animation is initiated.

(a)



(b)

Figure 4-20: Football player KeyFrame-based electrochromic display: (a) final prototype and (b) electronic circuit.

In Figure 4-21 is presented the circuit diagram required to operate the display. An Atmel ATMega328 8-Bit microcontroller was used as control unit for the electrochromic display.

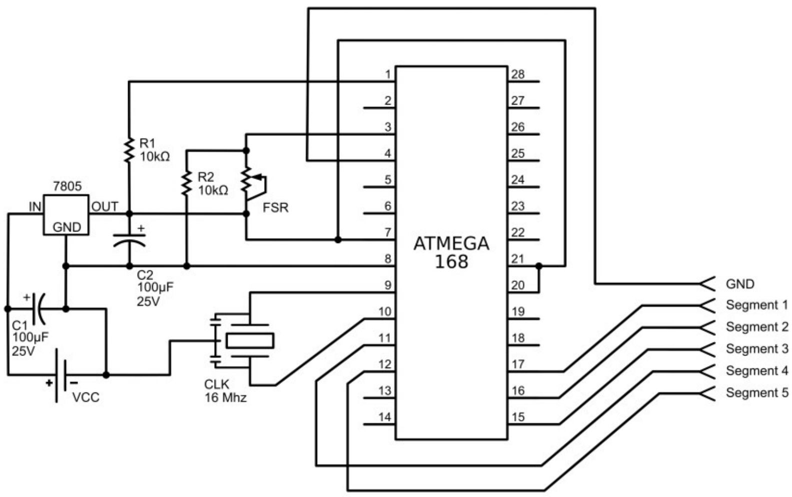


Figure 4-21: Circuit diagram of the “Football Player” electrochromic display control unit.

The circuit could be further optimised (or minimised) by using a microcontroller with less I/O lines, namely the Atmel ATtiny84 microcontroller, with a configuration to operate without requiring the external clock. The choice of both the ATMega328 microcontroller and the ATtiny84 microcontroller as preferable control units for the electrochromic display is related, first, due to their compatible with the Arduino programming environment, and second, for being both common microcontroller solutions used in the makers' communities. The source-code required to operate the display is provided in Annex F.

This type of approach is mainly suitable for convey a limited sense of movement, and there must be no overlapping between the various keyframes. Even so, the effects achieved can be quite entertaining. Possible applications include its use as a mimic of current animated neon signs.

4.2.3 Type II-A: Plain segment-based dynamic image

Type II-A electrochromic displays are segmented displays, based on the direct addressing method, in which the segments have the form of a simple geometric figure, normally of uniform shape and size. The visual content is updatable dynamically but predefined to an array of elements based on the graphic pattern and arrangement of the segments. The typical example, as seen in section 4.1.2.1.1, is the seven-segment configuration for displaying decimal numerals (Figure 4-22a). Each segment represents a picture element that can be individually switched *on* and *off* to give the appearance of the desired number. The segments can follow various designs (e.g. can have the shape of hexagons, trapezoids or rectangles) as well as have different spatial arrangement (e.g. segments can be arranged in an oblique position or have different sizes) according to the graphic pattern desired.

The seven-segment configuration can be extended to show both letters and numerals, i.e. to be capable of a full alphanumeric representation, through the implementation of either a fourteen-segment (Figure 4-22b) or sixteen-segment (Figure 4-22c) layout arrangement. However, as these layouts implement a higher number of segments, an increased number of drive circuits are required to operate them.

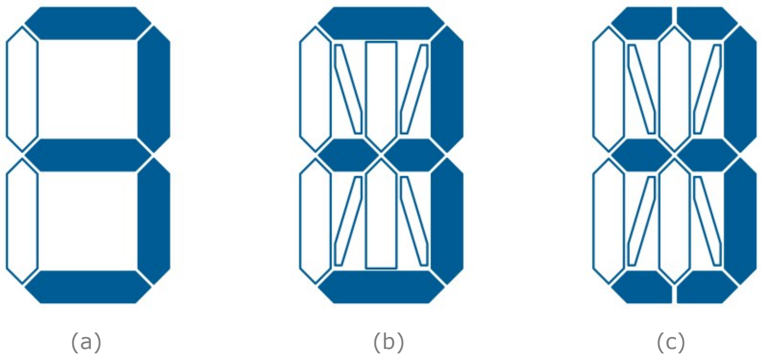


Figure 4-22: Layout configuration of a a) seven-segment, b) fourteen-segment, and c) sixteen-segment display.

Figure 4-23 illustrates a seven-segment electrochromic display reproducing the decimal numerals. The sequence of images illustrates the visual content this type of display can have, at the same time demonstrating the dynamic nature of the content based on a pre-determined configuration.

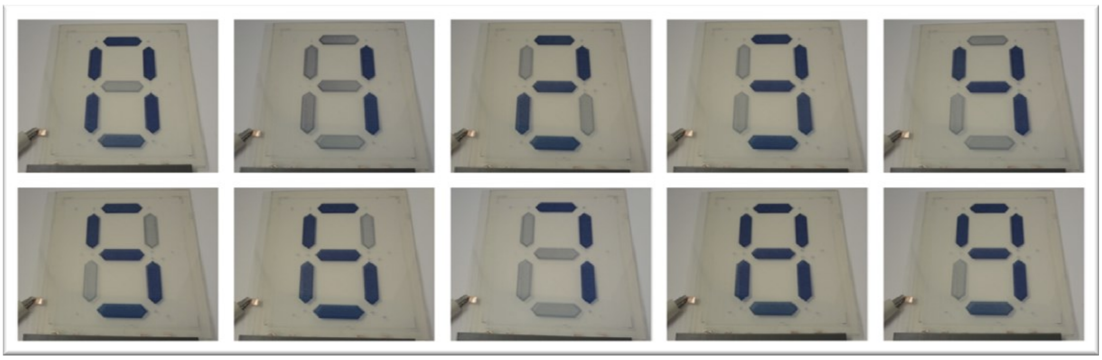


Figure 4-23: Example of a type II-A electrochromic display - Seven-segment display.

Beyond the typical seven-segment configuration, the use of segments is also common in a bar graph configuration, having applicability, for instance, as a battery charge indicator. Table 4-3 summarises the most common configurations in segmented displays.

Table 4-3: Common layouts for segmented displays.

Layout Configuration	Visual Pattern
7-Segment	Hexadecimal digits: Decimal numbers (0 to 9) and letters from A to F.
9-Segment	Decimal numbers (0 to 9) Basic alphanumeric characters
14-Segment	Standard ASCII characters (codes from 0 to 127).
16-Segment	Standard ASCII characters and table of extended codes (codes from 0 to 255).
n-Segment Bar Graph	Solid state level indicator

Other layout configurations and graphic patterns for the segments are possible, depending on the application and the information being conveyed. In Figure 4-24 is demonstrated a segmented display, following an approach different from the ones described above, where segments have a circular shape. The goal was to imitate the function of a dice and hence, have an abstract representation of the numbers from one to six based on dots. Like in the previous example of the seven-segment display, the visual content is dynamic but limited on the configuration of the segments.

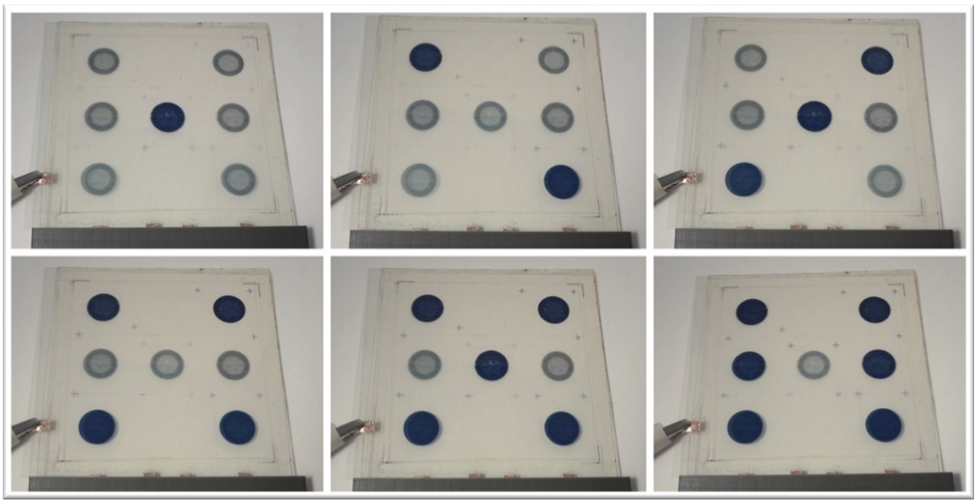
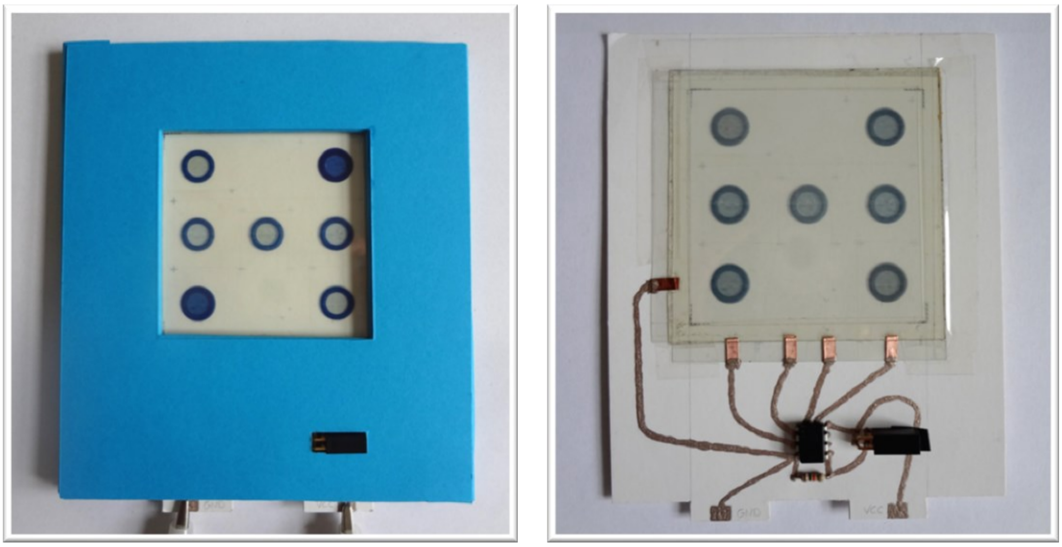


Figure 4-24: Another example of a type II-A electrochromic display – Digital dice.

Figure 4-25A shows the final functioning digital dice that you can "shake" to generate a random number. It was sought out to create the electronics invisible by blending it with the cardboard base and keeping the circuitry as thin and flexible as possible. Figure 4-25B shows the electronic circuit required to operate the device, in which the conductive traces are made with copper-based conductive ink. The display is operated based on a four-driver system: segments are activated in pars of two, in opposite positions, with the exception of the central segment. An Atmel ATtiny85 microcontroller was used to operate the display (see Annex G for the source-code used to operate the display). Figure 4-26 illustrates the circuit diagram required to operate the display.



(a) (b)

Figure 4-25: Digital dice electrochromic display: (a) final prototype and (b) view of the electronic circuit.

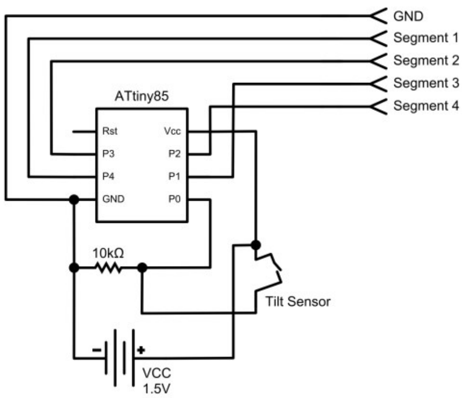


Figure 4-26: Circuit diagram of the "Digital Dice" electrochromic display control unit.

Segmented displays represent an advance in relation to type I-A and type I-B displays given that its content can be adapted to different situations to present specific information (even if limited to the arrangement of the segments). As a result, its content can convey different messages.

4.2.4 Type II-B: Multi pictograph-based dynamic image

In a type II-B configuration, each picture element consists of a pre-determined pictogram instead of an abstract geometric figure. Hence, every segment or pixel of the display has a distinctive meaning, conveying a specific message based on its visual resemblance to a physical object or event. As long as the receiver understands the simplified representation, pictograms are an efficient system for transmitting information in a simple, clear and semantically unambiguous approach whilst facilitating worldwide understanding. Pictograms are incredibly potent at transcending language, with its meaning being equally understandable to a multitude of people, independently of their idiom, culture and even literacy. Naturally, the implementation of standard sets of pictograms internationally agreed facilitates its multi-cultural understanding. Road signs and chemical hazard symbols are an example. Figure 4-27 illustrates various pictograms whose meaning transcend the barrier of verbal communication.

Figure 4-27: Example of various pictograms. Source: (Uebele, 2007).

The use of pictograms as message system is rather widespread these days, not only to inform but also to translate an advice, order or obligation. They are present in various environments from train stations and airports to hospitals, museums, exhibitions and business. Its applicability in electrochromic displays enables the creation of dynamically updatable visual communication systems such as dynamic signage systems and dynamic way-finding systems. Home applications intend to convey simple and clear information are another possibility.

In Figure 4-28 is illustrated a simple home application based on a direct addressing, three-segment pictographic electrochromic display. The pictorial components of the display (Figure 4-29) represent different streams of municipal solid waste and the application operates as a simple reminder service for household waste and recycling collection. In countries where the municipal solid waste is collected at a regularly scheduled day based on a door-to-door service, such as in Belgium, Italy, and some regions of Spain and the United Kingdom, remembering the correct days of collection can be a problem. Hence, the application visually reminds the user whenever is the correct collection day for each specific type of municipal solid waste by activating the respective icon. The source-code required to operate the display is provided in Annex H.

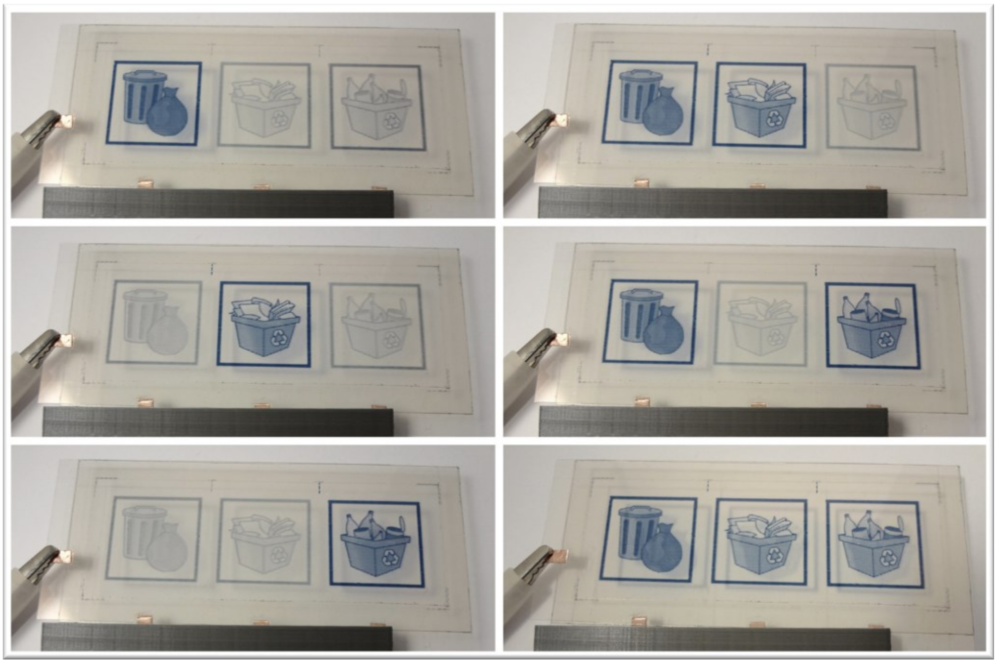


Figure 4-28: Example of a type II-B, segment-based electrochromic display – Waste reminder.

Figure 4-29: Municipal solid waste icons. From left to right: household waste, paper and cardboard recycling, mixed plastic and metal recycling.

Figure 4-30 depicts the circuit diagram of the display. An Atmel ATtiny85 microcontroller is used as control unit for the electrochromic display. The device could be further improved by implementing a real time clock module such as the DS1307 RTC to keep track of the time elapsed (though the circuit would be more complex).

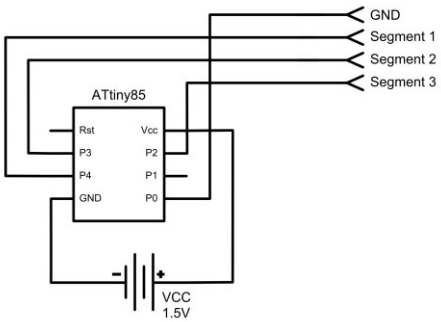


Figure 4-30: Circuit diagram of the “Waste Reminder” electrochromic display control unit.

Figure 4-31 illustrates another simple application based on a pictographic electrochromic display. The system was designed to visually present the weather conditions in a pre-determined city using a fixed set of weather forecast icons. In this case, the list of icons was limited to four basic weather statuses: sunny, cloudy, rainy, and snowy (Figure 4-32). A more comprehensive set of icons could be implemented by extending the display passive-matrix architecture to accommodate more rows and/or columns.

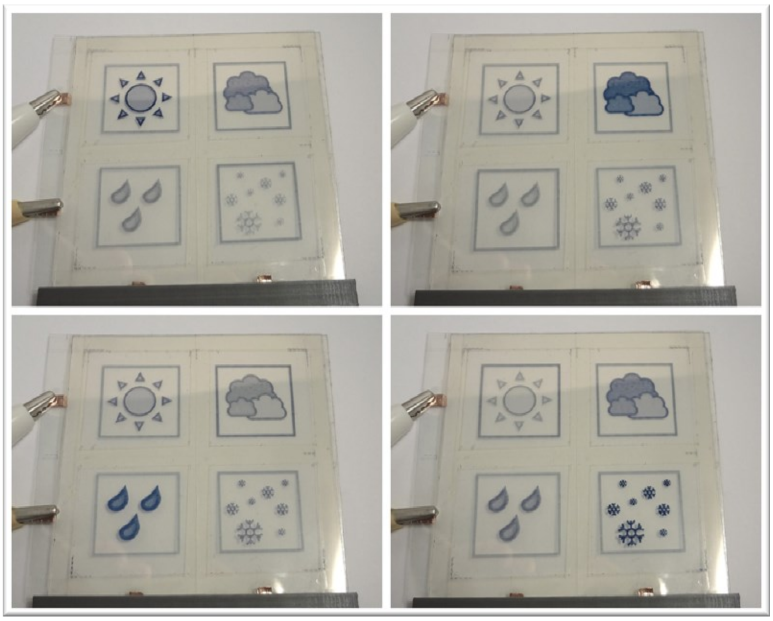


Figure 4-31: Example of a type II-B, matrix-based electrochromic display - Weather station display.

Figure 4-32: Weather forecast icons. From left to right: sunny, cloudy, rainy, and snowy.

The functionalities of type II-A and type II-B displays can be complemented, as seen in various segmented LCD screens, by combining the visual features of both types of display, enabling the presentation of both multi-character elements and pictogram icons. Figure 4-33 schematically illustrates this concept. It combines two seven-segment arrangements for showing the temperature values with a four-segment pictographic arrangement for displaying the weather condition in a similar way as seen in the previous example. As seen in type II-A, the content of type II-B displays can be changed (to a certain extent) to convey different messages. The particularity resides in the information context having to be defined beforehand.

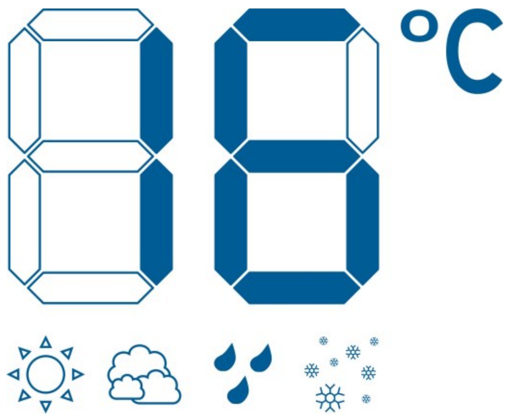


Figure 4-33: Illustration of a multi-character/pictographic segmented display.

4.2.5 Type III-A: Pixel-based dynamic image

A Type III-A configuration allows the creation of dynamic visual content that it is not limited to a pre-determined set of visual elements. The architecture follows a matrix structure in which the pixels are arranged in rows and columns, and have an abstract shape, typically of a square or a circle. The visual content is formed by individually activating the pixels so to produce the images and animations intended. The method used to drive the display can be either based on a passive-matrix or active-matrix addressing scheme.

Since the control of the visual content is done at the pixel level, the number of pixels available and thus the size of the matrix (number of rows and columns) greatly influence the visual potential of the display. A higher pixel density results in higher resolutions and more freedom in terms of visual content. Even so, a matrix display formed merely by five rows and three columns is already capable to reproduce the decimal numbers and a limited set of letters whilst a matrix display with seven rows and five columns is capable to reproduce all alphanumeric characters (Figure 4-34). Such matrix structure is as well capable to reproduce various simple pictograms and animations as illustrated in Figure 4-35.

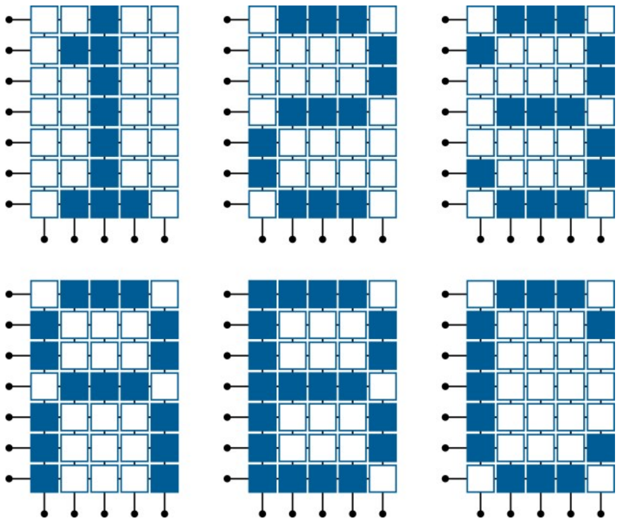


Figure 4-34: Illustration of a 7x5 matrix configuration reproducing diverse alphanumeric characters.

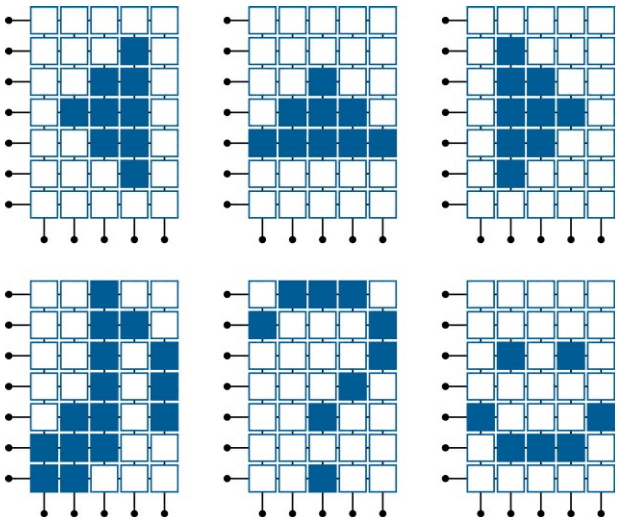


Figure 4-35: Illustration of a 7x5 matrix configuration reproducing diverse pictograms.

The display content can as well be manipulated at the level of pixel to engender the perception of motion. An example is the continuous movement of characters from a position to another, giving the illusion of scrolling text.

Beyond the possibility of unrestrictedly manipulate an image object, pixel-by-pixel, and create continuous animations, a type III-A architecture allows as well the implementation of interactive visual simulations. Individual pixels can be viewed as simple elements of a system that evolve through time and space following behaviour and interaction rules. Moreover, instead of using abstract geometric figures as pixels, pictorial images can be employed to represent the elements in the simulation.

4.2.6 Type III-B: Multi-concept pixel-based dynamic image

Type III-B configuration has a matrix structure similar to type III-A, with the difference that each pixel does not necessarily has the same geometric shape and more importantly, does not conveys the same information. The overall idea is that each pixel represents a pictorial entity that can have a specific meaning. This approach also intends to give low resolution matrix-addressing electrochromic displays a new level for presenting visual information. Likewise in type III-A configuration, the method used to drive the display can be either based on a passive-matrix or active-matrix addressing scheme. Figure 4-36 briefly illustrates the concept of using pictorial images as pixels. In this example, each pixel represents a living organism that can either be alive or dead, in an analogy to the Game of Life (see Box 5-1).

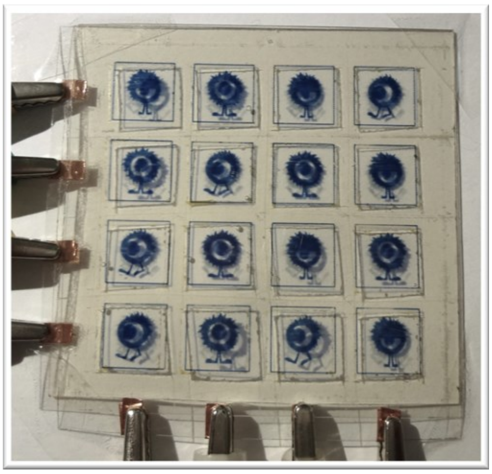


Figure 4-36: Example of a type III-B electrochromic display in which the pixels are pictorial images representing a living organism.

Indeed, the distinctive aspect of a type II-B electrochromic display is the fact that each pixel not only defines the overall visual content but as well can represent by itself specific information. Pixels (or group of pixels) have a distinctive meanings associated to them that overall yield a specific visual content. This idea can be further expanded by returning to the briefly presented notion of interactive visual simulations. If individual pixels are considered as simple elements of a system, they can very well represent different elements of that particular system and depending of the characteristics associated to each element or group of elements they can or cannot interact with each other. Figure 4-37 illustrates this concept. In the mock-up example presented, each pixel represents an individual element with a very specific behaviours (they represent different terrains types) in a simulation environment (the matrix). Depending on the behaviour and interaction rules applied to the simulation environment, the various elements (pixels) are affected in a different manner. If we consider, for example, the simulation of a forest fire, the elements representing forest areas and vegetation are going to be the main mean of spread for the fire whilst areas representing open water are considered as barriers. In turn, if we consider the simulation of a tsunami event, elements representing open water areas are the main mean of propagation of the event, whilst mountain areas are considered as barriers. In chapter 5 this notion is further explored, being proposed the implementation of cellular automata formulations to simulate the evolution of such systems.

Figure 4-37: Mock-up example of a type III-B configuration for a pictorial simulation environment (each square represents a pixel of the display).

4.3 Concluding remarks

This chapter addressed in detail the assembly of electrochromic displays, explaining the different components that compose one as well as the chemistry that governs the operation of such devices. The electrochromic devices manufactured for the purpose of this thesis used PEDOT:PSS as the electrochromic material, and were assembled according to the following configuration:

$$\langle\, (\, PET \mid ITO) \mid PEDOT{:}PSS \mid\mid Electrolyte \mid\mid PEDOT{:}PSS \mid (\, ITO \mid PET\,)\,\rangle$$

The choice of PEDOT:PSS as the electrochromic material was mainly because of its proprieties, in particular, its high chemical stability in normal conditions; its high transparency in thin, oxidised films; and its low electronic bandgap; as well as due to the fact that it can be processed from a water emulsion and easily deposited in various substrate, both rigid and flexible, by means of inkjet printing.

Design configurations regarding the control of individual picture elements in electrochromic displays were also tackled. The three main addressing methods explored were: (1) direct addressing, (2) passive-matrix addressing, and (3) active-matrix addressing. Each method has its advantages and challenges. The selection of one method over other influences the architecture and construction of the display, as well as the functioning of the microcontroller responsible for operating the display. The implementation of a particular addressing method in an electrochromic display is closely related to its desired application, the information content to be displayed and the production cost. The display area and the response time can also have influence in the choice.

Direct addressing is essentially convenient for display applications where there are only a reduced number of elements that have to be activated (e.g. alphanumeric displays). Given that in direct addressing displays, each picture element (commonly called segment) requires an individual connection, increasing the number of segments to further increase the information content of the display can significantly raise the number of connections to a point where managing all becomes too complicated, ultimately becoming unpractical due to the high cost of using so many drivers and the absence of space between elements for the higher number of connections.

Passive-matrix addressing solved the limitations of direct addressing in present high information content by moving to a scheme composed by pixels arranged in a matrix with row and column electrodes. The novelty was in the picture elements, i.e. the pixels, being addressed by their row and column instead of being driven separately. Moreover, the simplicity of the scheme also made it very cost-efficient. However, it also raised new issues that ended up limiting its practical applicability. As the size of the passive-matrix increased, the image contrast ratio decreased and the response time slowed down. Also, it was observed voltage leaks between neighbouring pixels in the same row or column electrodes, resulting in the colouration of undesirable elements (crosstalk effect).

Finally, the active-matrix addressing scheme appears as a solution to the problems associated with passive-matrix displays, such as the scanning limitations and crosstalk effect. The construction is similar to the passive-matrix, with pixels arranged in a matrix with row and column electrodes. However, active-matrix display have a thin-film transistor built into each pixel, at the cross point of the row and column electrodes. The TFT acts as a switch, precisely controlling the voltage of each pixel. The drawback of the active-matrix scheme when compared to the passive-matrix is mainly related to its higher cost and manufacturing complexity.

The different types of display architecture/content detailed in this chapter highlight how electrochromic displays can be used to present different levels of visual information. It intends to provide a clear view of the various possibilities offered and what can be expected to be achieved in terms of dynamic content. The complexity of the architecture of the different arrangements increases from type I to type III, likewise the potential for presenting dynamic information.

The dominant feature of type I displays is that content is pre-determined and limited to a fixed number of difference instances. In type I-A, the maximum number of instances is two whilst in type I-B the content is segmented to provide multiple instances of the same image. In turn, the particularity of type II displays lies in the content being updatable but limited to a fix pre-set of arrangements based on layout of the composing segments. In type II-A displays, the individually addressed segments are composed by abstract elements that can be combined to form numbers, letter or symbols whilst in type II-B, the segments are composed by individual pictograms, each one with a very specific meaning. Finally, in type III displays, the matrix structure of the display allow the creation of dynamic content through the differentiate control of the different picture elements. Type III-A represents the typical all-purpose matrix arrangement display, where it is possible to create multiple compositions and animations whilst the type III-B represents a arrangement where a specific meaning is associated to each individual picture elements that influences the overall visual content of the display. An envisaged application, addressed in detail in the next chapter, is the use of such arrangement as a means to display simulations based on pictorial elements performed by cellular automata models in real time.

Electrochromic displays are still in their infancy stage in terms of real word applications, and hence, experimentation in real world contexts is still necessary. The various examples advanced here intend not only to demonstrate the different types of architecture/content but as well provide insights for possible applications. The particular technical universe explored showed to be appropriate for making interactive devices. With the exception of Type III-B displays (which were only idealised), all examples were developed using off-the-shelf components and hardware. A key point was to allow for user participation in the creation of the devices themselves. Naturally, it would be interesting to see how the makers' communities would embrace and use this technology in their projects, considering the vast array of possibilities for customisation.

5 Computing with Simple Programs

5.1 The world of cellular automata

In the late forties, the mathematician John von Neumann was trying to formalise an abstract model of self-reproduction in biology. Following a suggestion from his friend and fellow mathematician Stanislaw Ulam, of using a discrete system for creating a reductionist model of a self-replication machine (Mitchell, 2009: 123; Wolfram, 2002: 876), von Neumann envisaged a checkerboard look-like system, in which each square was a finite state automaton. In this particular system, each cell could be in one of twenty-nine possible different states and would follow pre-determined rules specifically set up to create a perfect reproduction of any initial pattern placed on the cellular automaton lattice (see Neumann, 1966). Von Neumann's mathematical formulation of a self-replicating automaton was at the time a major advance in the field of artificial life, demonstrating that a) self-reproduction by machines was in theory possible and b) fairly simple rules could form complex patterns. Moreover, he demonstrated that the devised cellular automata could execute any computable operation. However, due to its complexity, Von Neumann rules were never implemented on a computer.

In 2002, Stephen Wolfram published a likewise acclaimed (see Rucker, 2003) and controversial (see Giles, 2002) book entitled "*A New Kind of Science*" (or NKS for short) (Wolfram, 2002). In it, Wolfram set out to study the behaviour of simple programs, in particular of cellular automata. The author argues that every process found in nature can be regarded as a computation[25] that has very simple, definitive, underlying rules. The core idea is that simple programs such as cellular automata, despite the simplicity of their underlying rules, can exhibit highly complex and random-looking behaviour, and somehow the same basic mechanisms responsible for this phenomenon in simple programs are present in the natural world producing the wealth of complexity perceived everywhere. Wolfram states that it is

[25] Computation is viewed here in a broader concept. It is regarded as the process of transforming inputs into outputs, where the inputs and outputs are states of the underlying system that supports the computational process.

remarkable how often this occurs, with natural systems frequently showing complex and random-looking behaviour that visually looks almost identical to what is seen in certain simple programs. As an example, he points out things like the patterns in the pigmentation of mollusc shells that seem to follow a one-dimensional cellular automaton (Figure 5-1), or the snowflake formation process that can be remarkably reproduced by a two-dimensional cellular automaton (Figure 5-2). Hence, these simple programs provide a new framework for understanding the immense complexity that characterise complex systems[26]. Instead of searching for the mathematical equation that try to represent the mechanisms by which certain phenomenon is produced, one should look for the simple program which reproduces the striking features observed in that specific system.

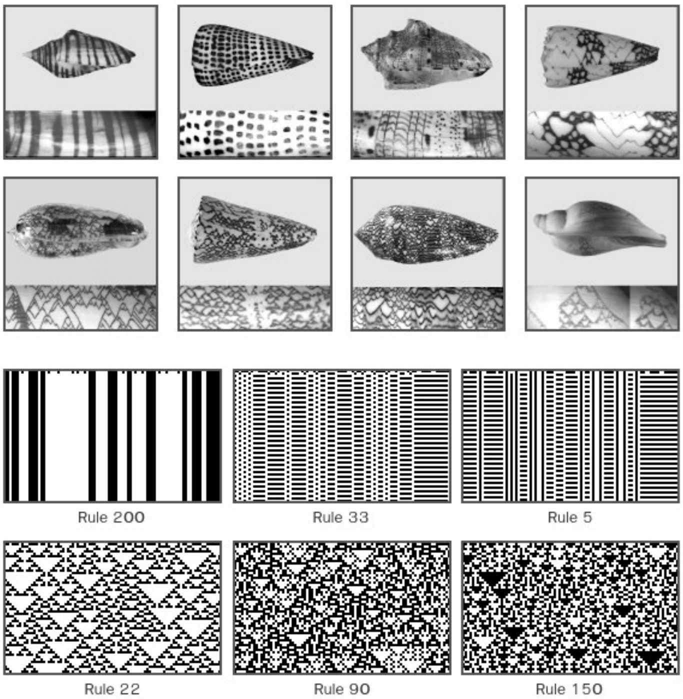


Figure 5-1: Comparison between examples of pigmentation patterns on mollusc shells (top images) and the evolution of specific one-dimensional cellular automaton rules (bottom images). Source: adapted from (Wolfram, 2002).

[26] For a comprehensive analyses on the subject of complexity and complex systems, see for instance (Mitchell, 2009; Waldrop, 1993).

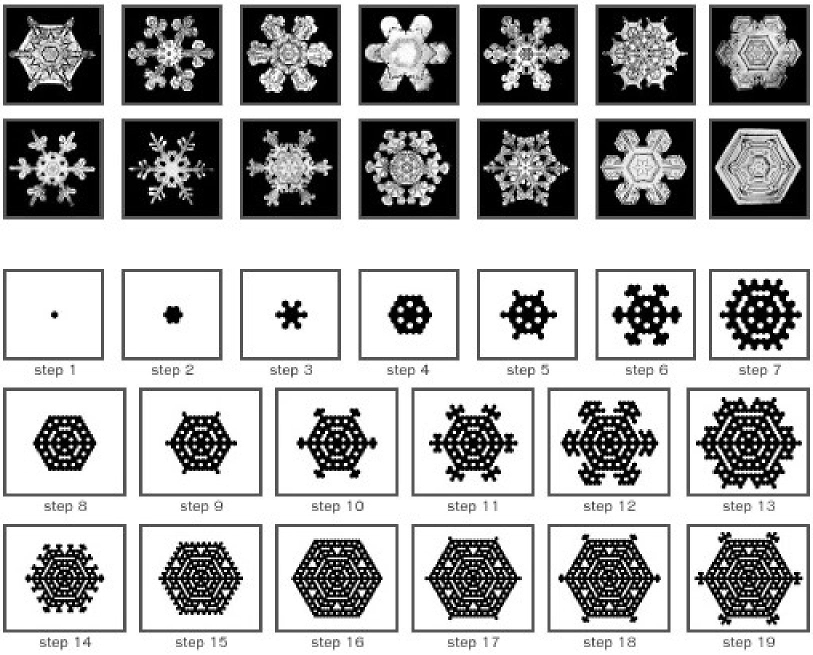


Figure 5-2: Comparison between examples of typical snowflakes patterns (top images) and the evolution of a two-dimensional cellular automaton that remarkably reproduces the basic snowflake formation process (bottom images). Source: adapted from (Wolfram, 2002).

Moreover, Wolfram mentions that the phenomenon of complexity is quite common in nature, and that the sophistication of the computations actually being performed are in fact equivalent. Rather than some complex systems being simpler than others, Wolfram feels that nearly all of them are of equal and maximal complexity. When a natural phenomenon is not obviously simple, it means that it is not only complex, but of maximal complexity. From a computational point of view, every single one of these systems, despite their great differences in structure, is ultimately able to support universal computation[27]. The immediate suggestion is that the ability to support universal computation is thus very common in nature and rather readily attainable. Once a system passes a certain computational threshold, in effect a very low one, it becomes universal. It means as well that there is an upper limit on the complexity of possible computations in nature and elsewhere. That is, there is nothing more complex than what can be computed by a universal computer. However, this also implies that there is not going to be a simple and

[27] A computation is universal if it can emulate any other computation, i.e. if it is able to perform any other possible computation. Hence, given enough time and memory, any universal computer can simulate any other type of computer, or for that matter, any other device or system that processes information (Hillis, 1998; Rucker, 2005). Most real-life computers are universal. Note, nonetheless, that there are certain things that are just simply impossible to computer (e.g. the halting problem (Turing, 1936)). See also (Harel, 2000; Rucker, 1987).

rapidly running computation that can emulate the natural world much faster that it happens (Rucker, 2005; Wolfram, 2002).

Wolfram approaches various fields of science from the perspective of simple programs, namely mathematics, physics, biology, fluid dynamics, artificial intelligence, artificial life, chaos theory, complexity theory, economics, consciousness, extra-terrestrial intelligence, and, of course, computer science. He goes to the point of speculating that there is a "*simple program which, if run for long enough, would reproduce our universe in every detail*" (Wolfram, 2002: 465) - the ultimate model for the universe whose computations are the source of everything that exists[28]. It is worth mentioning that the ideas and claims presented in "*A New Kind of Science*" are far from consensual, being the target of scrutiny by various scholars (see (Bailey, 2002; Kadanoff, 2002; Kurzweil, 2002; Mitchell, 2002)).

Considering the framework of this thesis, the core idea to retain is the inherent potential of simple programs such as cellular automata in performing complex computations in a non-traditional way. Systems with extremely simple underlying rules can indeed produce complex outcomes and once a certain threshold for complex behaviour has been reached, making the underling rules of a system more complex does not ultimately lead to more complex overall behaviours. From an aesthetical point of view, cellular automata provide an interesting way to produce new forms of images that are capable of changing interactively (spatial images).

5.2 The basics behind cellular automata

Cellular Automata (see for instance, (Toffoli and Margolus, 1987; Wolfram, 1983, 2002)) are discrete dynamical systems consisting of a uniform grid (or lattice) of simple identical components, commonly called cells. The overall structure can be viewed as a parallel processing device since the system has no central controller and the communication between constituent cells is limited to local interaction. Time advances in a succession of discrete steps and at each time instance each individual cell exists in a specific state from a finite number of overall possible states (for example, in the simplest possible cellular automata

[28] The idea that the universe might be like a cellular automaton was in fact originally suggested by Konrad Zuse as early as 1967 (Schmidhuber, 2002; Zuse, 1970). Likewise, Edward Fredkin theorised the same concept, though some years later, before becoming acquainted with the work of Zuse (Fredkin, 1992; Wright, 1988).

configuration two states are considered: *on* and *off*). The state of each individual cell over time is determined based on the states of its local neighbour's cells at the preceding time step according to the application of a finite set of pre-determined transition rules (also known as local rules) to the system. The dynamics of the system results from repeatedly implementing the transition rules to every cell on the grid. The rules can be represented in numerous different ways, from a set of IF-THEN statements to a formula or a lookup table. Totalistic rules are those in which the new state of a cell depends only on the average of the previous states of its neighbours as well as on its own previous state. The best known two-dimensional totalistic cellular automaton is the Game of Life (see Box 5-1).

Normally, the transition rules are applied simultaneously to every cell, i.e. there is a global clock that dictates the pace of all local processes in the system, and are identical for all cells. Even so, there are examples of asynchronous cellular automata (Capcarrere, 2002; Cornforth et al., 2003; Ingerson and Buvel, 1984; Nehaniv, 2003; Schönfisch and de Roos, 1999; Zielonka, 1987)[29], where the state of each cell is updated sequentially in such a way that the new state of a cell influences the calculation of the state of neighbouring cells. The update sequence in which the cells are considered can be described as either random or ordered. Common employed approaches include (Cornforth et al., 2005): (a) the *clocked scheme*, where each cell has its own independent timer, so that updating is autonomous and proceeds at different rates for different cells; (b) the *self-sync scheme*, similar to the clocked scheme, but the period of each timer is adjusted after an update so as to more closely match the period of other cells in its neighbourhood; (c) the *cyclic scheme*, where at each time step a cell is chosen according to a fixed update order, which was decided at random during the initialisation of the system; (d) the *random independent scheme*, where one cell is randomly selected for update at each time step; and (e) the *random order scheme*, where all cells are updated in a random order at each time step. Likewise, there are examples of cellular automata where the transition rules are not identical for every cell (i.e. each cell may contain a different rule). These are commonly called hybrid or non-uniform cellular automata (Dennunzio et al., 2012; Dogaru, 2009; Sipper, 1994). In these particular cases of cellular automata, there is a collection of transition rules that are applied according to the position of the cell.

One of the most fundamental properties of cellular automata is the dimensional arrangement of the lattice on which the system is computed. In theory,

[29] It is argued that the synchronous approach is not realistic from a biological point of view since there is no accurate evidence of global synchronization in nature (Capcarrere, 2002; Cornforth et al., 2003).

it can be in any finite number of dimensions (*d*-dimensional). The simplest possible arrangement consists of a one-dimensional array (Figure 5-3a) (Wolfram, 1984a). Two-dimension (Figure 5-3b) (Packard and Wolfram, 1985) and three-dimension (Figure 5-3c) (Davies, 2009; Reiter, 2011) spatial arrangements are as well common in literature. In effect, cellular automata initially appeared in terms of a two-dimensional grid of cells. Three-dimensional arrangements have been less explored, possibly because of the higher computational power required to render the three-dimensional lattices (which until some years ago was not available), and the vastness possibilities of the topic.

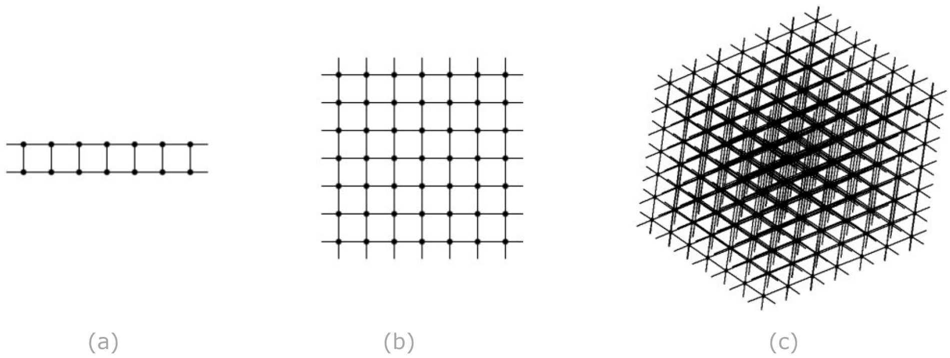


Figure 5-3: Common spatial arrangements of cellular automata elements in a) one-dimension, b) two-dimension, and c) three dimension lattices.

The grid can have either a null or periodic boundary. In the case of a null boundary, the cells on the edge of the grid are assumed to have a void dependency (i.e. a logic value of “0”) or alternatively a fixed value. A periodic boundary is one in which the grid is considered to be folded, with cells on opposing edges functioning as neighbours.

Individual cells are commonly depicted as squares though in two-dimension lattices, triangular and hexagonal cells are as well recurrently implemented (Figure 5-4).

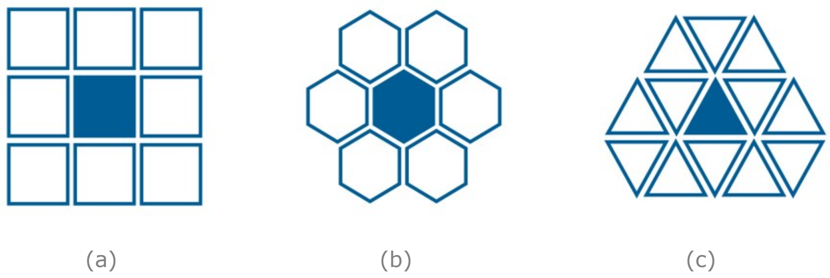


Figure 5-4: Example of commonly used cell tilling: a) square tilling, b) regular hexagon tilling, and c) regular triangle tilling.

The set of cells that define the neighbourhood of a particular cell must also be specified. Since there are no restrictions on which cells can be considered, a wide variety of neighbourhood configurations can be implemented. In one-dimension cellular automata, it is common to define the immediate adjacent cells on the left and right of a particular cell as its neighbours. Naturally, it is possible to extend this configuration to include a higher number of adjacent cells. In two-dimension cellular automata, the most know types are the Von Neumann neighbourhood, which consists of the four orthogonally adjacent cells, (Figure 5-5a) and the Moore neighbourhood, which consists of the complete eight surrounding adjacent cells (Figure 5-5b).

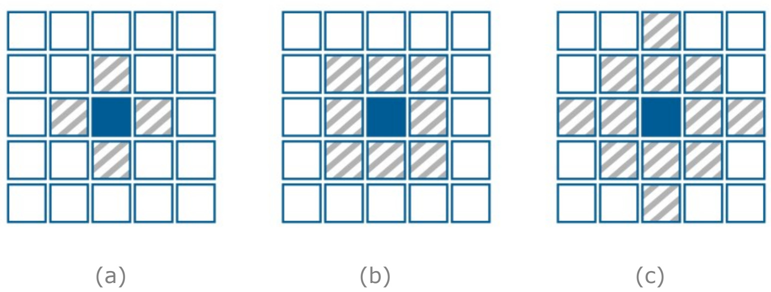


Figure 5-5: Classical neighbourhood of two-dimension cellular automata: a) von Neumann neighbourhood, b) Moore neighbourhood, and c) a combined Moore neighbourhood with an extended von Neumann neighbourhood.

Box 5-1: John Conway's "Game of Life".

Game of Life

The "Game of Life", or simply "Life", is a two-dimension cellular automaton that was devised by the mathematician John Conway. It is the most widely known example of cellular automata, having reach an immediate popularity when it was first mentioned by Martin Gardner in his monthly column "Mathematical Games" of the October 1970 issue of Scientific American (Gardner, 1970). The overall excitement was primarily due to the fascinating patterns and intricate behaviours that emerged from the very simple rules behind the computations.

In the Game of Life, individual cells can have either the value of 0 or 1 bit, denoting respectively if the cell is dead or alive. It is a totalistic cellular automaton meaning that the new state of a cell depends only on the average of the previous values of its neighbours as well as on its own previous value. The neighbourhood of each cell consisting of the eight surrounding adjacent cells (Moore neighbourhood). Likewise cells are described as being alive or dead, the Game of Life rules (Figure 5-6) are as well commonly phased in terms of life processes:

- A dead cell becomes alive at the next time step when that cell has exactly three live neighbours (birth). Otherwise a dead cell stays dead.

- A live cell with exactly two or three live neighbours stays alive at the next time step (survival). Otherwise it dies of loneliness (cell with fewer than two live neighbours) or overcrowding (cell with more than three live neighbours).

The Game of Life balances these tendencies, making it difficult to figure whether a pattern will die out completely, form a stable population, or grow forever.

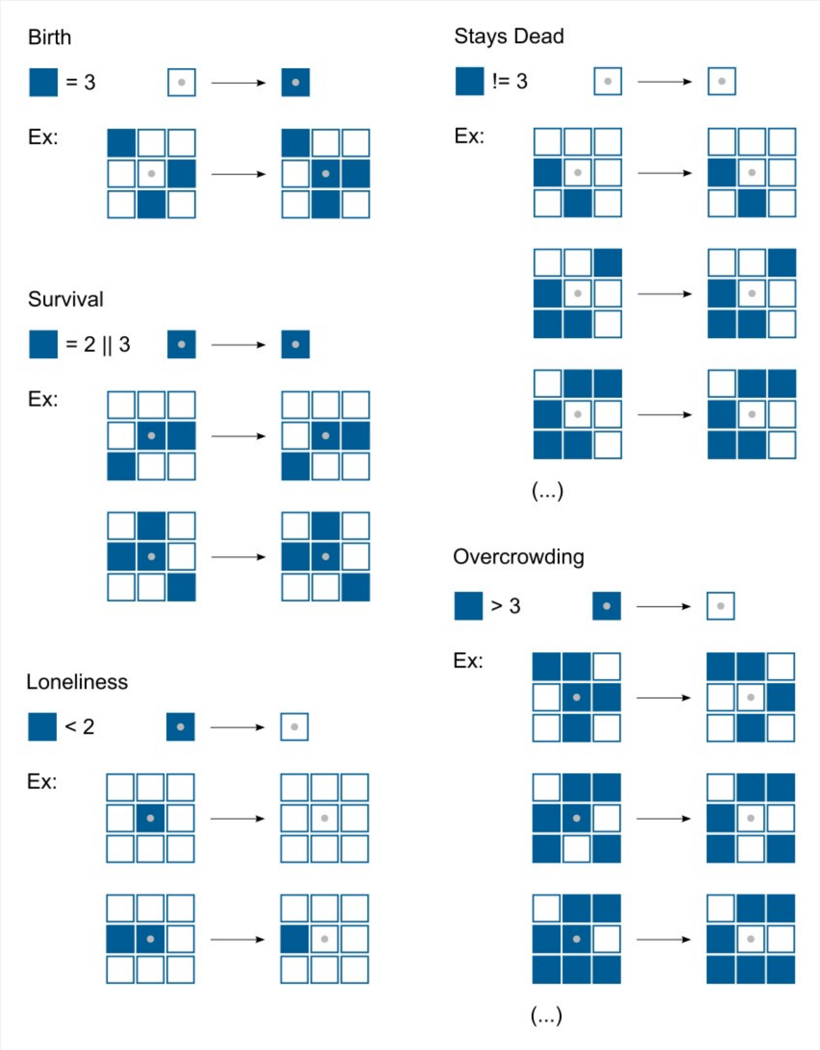


Figure 5-6: The Game of Life rules.

Depending on the initial configuration of the cells, particular patterns of repetitive or other interesting behaviour can originate from the space-time evolution of the Game of Life. For example, William Gosper and five fellow hackers at MIT were the first to prove the existence of a simple initial configuration that grows without limit (Rucker, 2005: 64). The configuration evolves into a “glider gun” and after producing its first glider, every 30 steps, it originates a new one. This glider gun is still the smallest one known. Other interesting types of patterns exist in the Game of Life, including still lifes (patterns that do not change from one generation to the next - Figure 5-7), oscillators (patterns that repeat themselves after a fixed number of generations - Figure 5-8), and "spaceships" (patterns that translate themselves across the board - Figure 5-9).

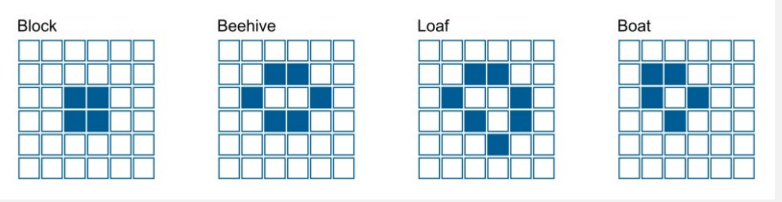


Figure 5-7: Example of common still lifes in the Game of Life.

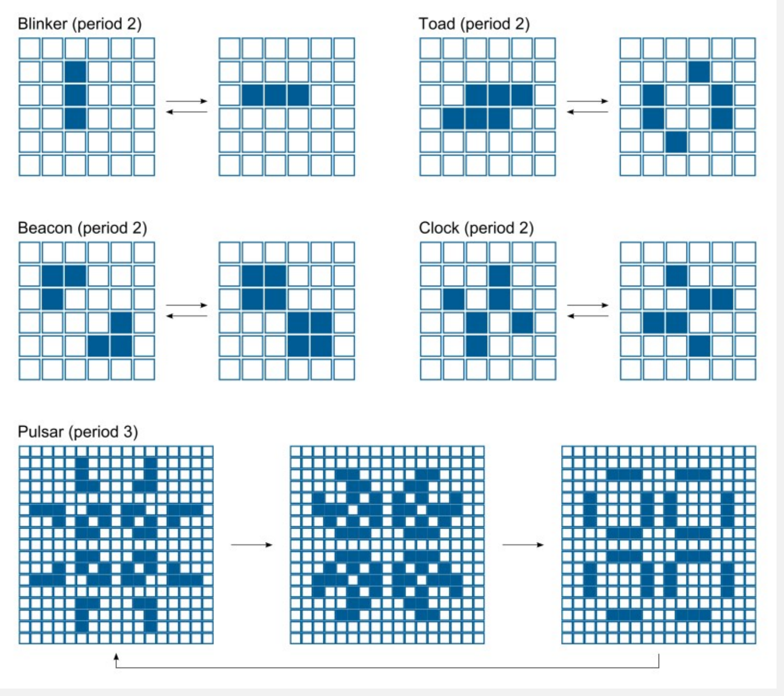


Figure 5-8: Example of common oscillators in the Game of Life.

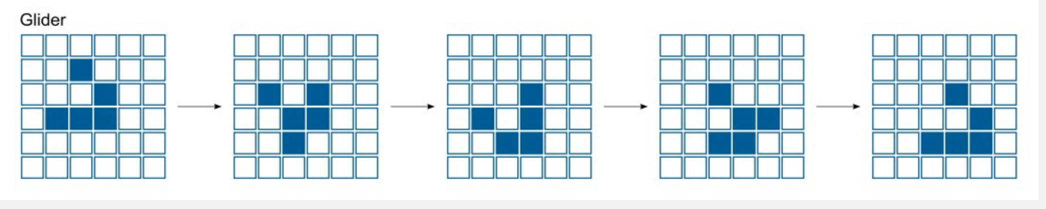
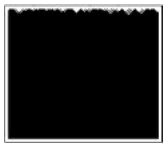
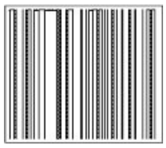
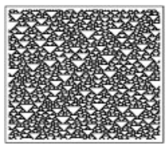
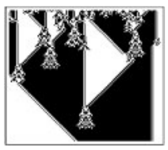


Figure 5-9: Example of a "spaceship" moving in the Game of Life.

It has been proved as well that the Game of Life is capable of universal computation (Elwyn Berlekamp et al., 2001) by demonstrating how glider guns, gliders, and other structures could be assemble so as to carry out universal logical functions (i.e. *and*, *or*, and *not* operations). For an in-depth analysis of the Game of Life, see for instance (Adamatzky, 2010).

Depending on the overall behaviour that a cellular automaton computation exhibits through time, it is possible to categorize it in one of four basic computation classes (Wolfram, 1984b). The different classes enable different levels of prediction of the outcome from particular initial states. Class one and class two computations are considered simple, while class three and class four present complex behaviour. Note, nonetheless, that the borders between the different computations classes are not always clear, namely between class three and class four. Table 5-1 describes each class. The examples provided relate to the evolution of elementary one-dimensional cellular automata from random initial conditions.

Table 5-1: Cellular automata basic classes of behaviour.

	Description	**Behaviour**	**Example**
Class 1	Initial conditions generate a uniform final pattern.	Simple	
Class 2	Initial conditions generate a repetitive or nested final pattern.	Simple	
Class 3	Initial conditions produce random-looking behaviour, although regular structures can be present.	Complex	
Class 4	Initial conditions generate a mixture of order and randomness: areas of repetitive or stable states are created as well as structures that interact with each other in complicated ways.	Complex	

In the last three decades, cellular automata have been studied and applied in a wide range of different fields of science. Examples include the use of cellular automata in:

- Modelling dynamic systems in nature and society (e.g. urban development (Barredo et al., 2003; Li and Yeh, 2000), earthquakes (Georgoudas et al., 2007; Nakanishi, 1990), oil spills (Nakano et al., 1998; Rusinovic and Bogunovic, 2006), forest fires (Karafyllidis and Thanailakis, 1997; Yassemi et al., 2008));

- Modelling physical systems (e.g. fluid dynamics (Frisch et al., 1986; Wolfram, 1986), gas behaviour (Boon et al., 1996; Zahedi Sohi and Khoshandam, 2012)),

- Modelling chemical systems (e.g. first- and second-order kinetic phenomena (Kier et al., 2000; Seybold et al., 1997), Belousov–Zhabotinsky reaction (Gerhardt and Schuster, 1989; Markus and Hess, 1990))

- Modelling biological systems (e.g. predator-prey dynamics (Droz and Pękalski, 2001; Pekalski, 2004), immunology (Chowdhury et al., 1990; Sieburg et al., 1990), cell grow (Alarcón et al., 2003; Ermentrout and Edelstein-Keshet, 1993), pigmentation formation (Gunji, 1990; Markus and Kusch, 1995));

- Idealizations of massively parallel, non-centralized computation (Hansen, 1993; Spezzano and Talia, 1999);

- Cryptography as generators of random numbers (Nandi et al., 1994; Seredynski et al., 2004).

- Animation (e.g. clouds (Dobashi et al., 2000; Miyazaki et al., n.d.) and water waves (Wang et al., 2003))

Comprehensive surveys highlighting the theory and applications of cellular automata can be found in (Ganguly et al., 2003; Kari, 2005; Kutrib et al., 1997; Mitchell, 1998).

5.3 Computing with pictorial entities

Most computations are primarily done based on numerical entities due to these being easily interpreted, written and manipulated (Nickerson, 1988). However, in our daily life, we mainly use linguistic and pictorial entities to describe the world. The use of linguistic (Zadeh, 1975) and pictorial (Camara et al., 1994) entities in computation allows the shift from strictly quantitative variables into more qualitative and distributed representations of reality. Indeed, pictorials entities can be a powerful and efficient mean to represent concepts and ideas, overcoming as well existing language barriers.

The matrix-based architecture of type III electrochromic displays makes them well-suited to displaying cellular automata. This section addresses how pictorial entities can be used in electrochromic displays to perform pictorial simulations of dynamic systems. The pictorial approach described herein follows the work developed by (Camara et al., 1994). In this methodology, pictorial entities include pictographs (realistic representations of real phenomena or objects), symbols (abstract representations of phenomena or objects), and (arbitrary) signs. The computation scheme is similar to that of the cellular automata approach. Hence, the simulation occurs in a space grid system that evolves through discrete time steps advances. Each pictorial entity is regarded as a single cell which has a predefined behaviour or behaviours. Interaction between pictorial entities is possible but limited to neighbourhood cells. The transition rules that compose the system are thus divided in behaviour and interaction rules (Table 5-2).

Table 5-2: List of pictorial transition rules.

Basic Rules		**Description**
Behaviour Rules	Movement	Displacement of a specific pictorial entity in the X-Y axis.
	Expansion	Increase in size of the tile set referent to a specific pictorial entity.
	Retraction	Decrease in size of the tile set referent to a specific pictorial entity.
	Decay	Elimination of a specific pictorial entity.
Interaction Rules	Attraction	One or both pictorial entities are draw close to the other.
	Repulsion	One or both pictorial entities are draw apart from the other.
	Neutralisation	Elimination of both pictorial entities when interaction occurs.
	Reproduction	Creation of a third pictorial entity when interaction occurs.
	Transformation	Replacement of one or both pictorial entities for another when interaction occurs.
	No change	Nothing changes when interaction occurs.

Source: adapted from (Camara et al., 1994; Nobre and Câmara, 1999).

The formulation of the rules follows a biological analogy making them especial adequate to model physical, chemical, biological, and environmental phenomena (Camara et al., 1990; Nobre and Câmara, 1999). Figure 5-10 illustrates the application of the behaviour and interaction rules in a two-dimensional spatial arrangement.

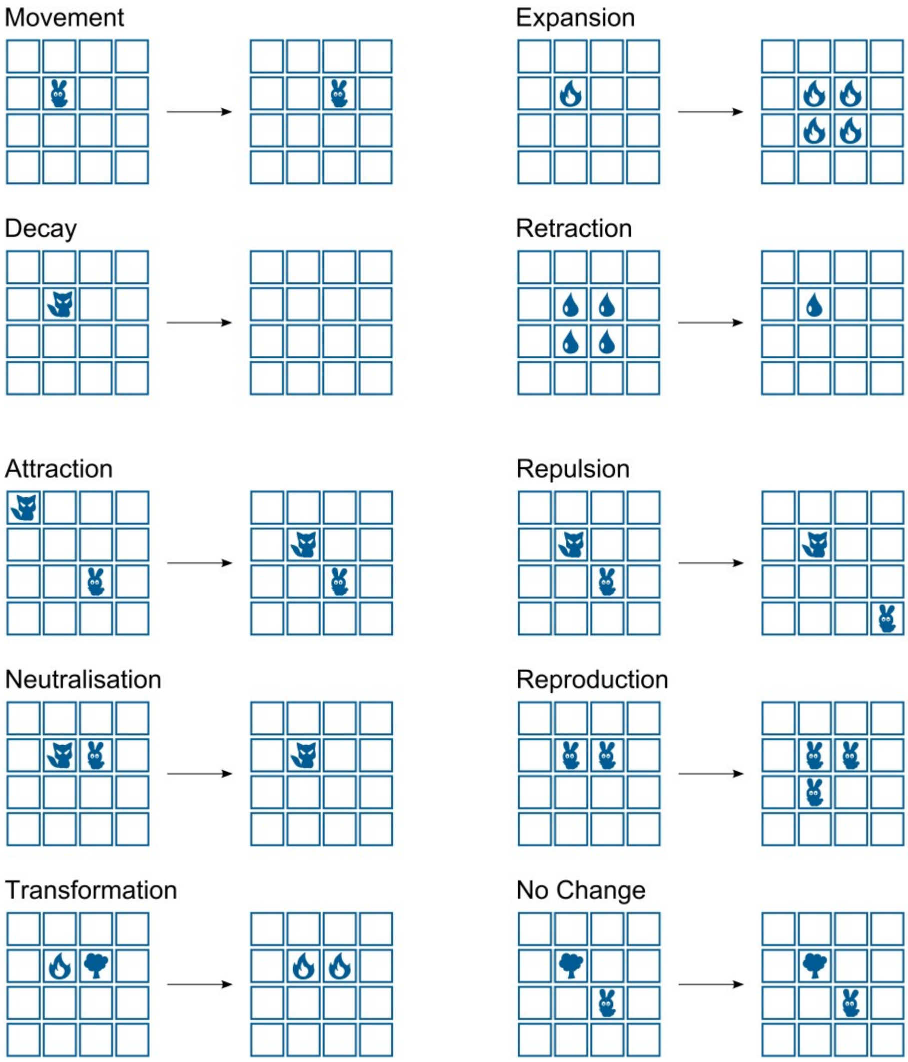


Figure 5-10: Exemplification of the pictorial simulation behaviour and interaction Rules.

5.3.1 Implementation in electrochromic display

The implementation of the pictorial simulation approach was envisaged in a multi-layer architecture based on a series of interconnected modules of stackable electrochromic displays (Figure 5-11).

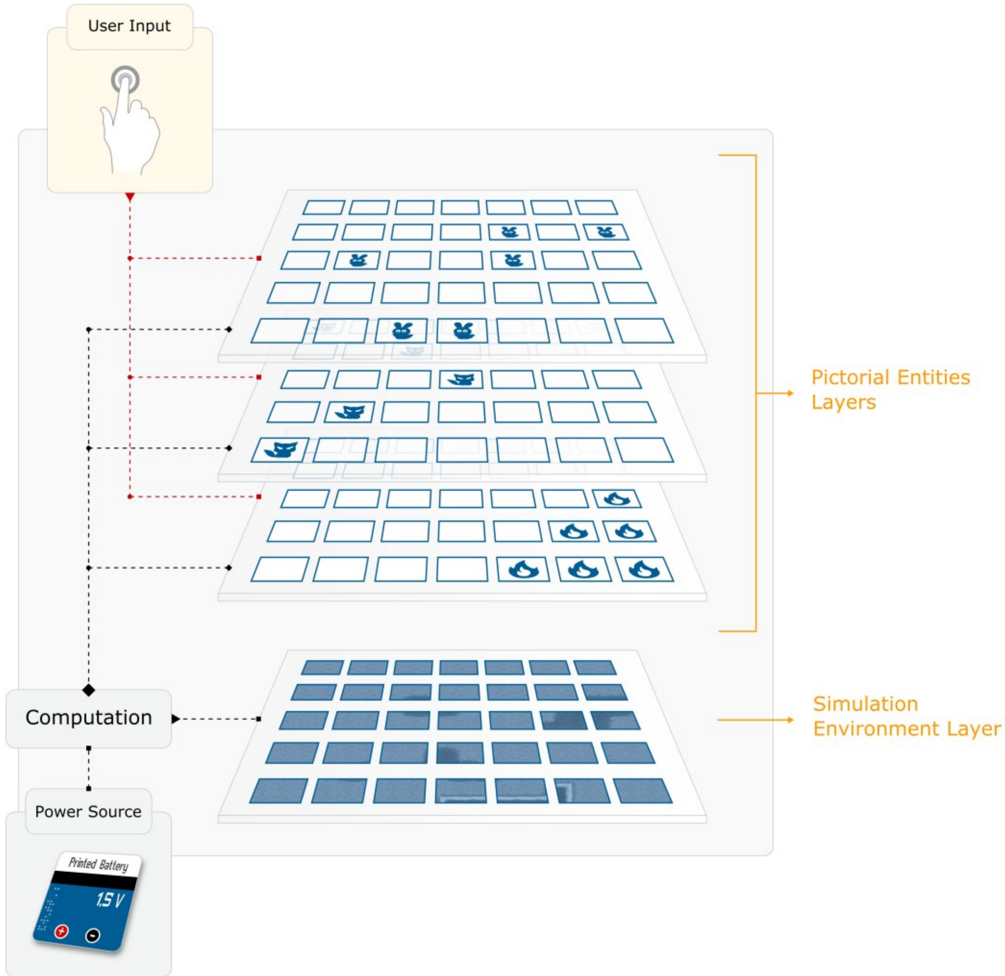


Figure 5-11: Architecture of the pictorial simulation environment.

The system is divided in a two-level hierarchical structure. The base level encodes the overall space and time structure of the system such as the type of boundary (cyclic or null), the type of neighbourhood (von Neumann, Moore or other), and the global clock (synchronous or asynchronous). It is formed by a single layer representing a particular simulation environment.

The top level provides the logic for the simulation. It encodes the specifications of the components that form the system. It is composed by one or more layers, each representing a specific pictorial entity with a pre-determined set of behaviour and interaction rules. Each layer can function independently from the others but also in interaction. Hence, when only a pictorial entity layer is implemented in the system, the simulation will be based essentially on the behaviour rules created for that specific pictorial entity. In contrasts, when multiple layers are stacked together, i.e. connected to one another, the simulation will be based both on the behaviour rules of each pictorial entity but also on the interaction rules. This allows the engineering of specific interactions between certain pictorial entities.

The system is idealised to allow the layers to be changed easily. Also, additional ones can be added to the system at any given moment, enabling new pictorial entities to be incorporated to the simulation. The crucial element of this architecture is that all information and instructions must be clearly defined and separated according to the hierarchical structure of the system. The system architecture was replicated in a computer simulation. The code diagram for system is provided in Annex I.

Pictorial simulation can be applied to various spatial simulation problems (Camara et al., 1990, 1994; Nobre and Câmara, 1999). As mentioned above, the formulation of the rules follows a biological analogy making them especially adequate to model physical, chemical, biological, and environmental phenomena. Typical uses include the modelling of forest fires, floods, oil spills, and predator-prey dynamics. From an educational application standpoint view it provides a tangible way for experimenting with complex systems (and acquire knowledge) using straightforward pictorial entities and intuitive transition rules.

5.4 Concluding remarks

Cellular automata are a fascinating type of computation. Despite of their underlying simple structure, cellular automata are capable of producing outcomes that are far from simple. One of the obvious attractions of cellular automata is the complex and often beautiful visual patterns that they can generate. The number of interesting configurations that can be made from these elements is immense. Other appeal of cellular automata is the potential to simulate different sophisticated natural phenomena. By using the type of rules embodied in cellular automata it is possible to capture a vast range of essential mechanisms in physical, chemical, and biological systems. The Game of Life is one of the best know cellular automata. It is characterised by generating a great number of structures such as oscillators and "spaceships" capable of producing interesting behaviour on the boundary between stability and chaos. With proper initial conditions it is also capable of simulating any given computation. In fact, there are cellular automata systems whose rules are simple enough to be described in a few sentences that are nevertheless capable of universal computation (e.g. rule 110 (Cook, 2004)).

The matrix-based architecture of type III electrochromic displays naturally suggests its use with cellular automata. A clear analogy can be made between the individual cells in cellular automata and the pixels in an image, with the particularity that the dynamic nature of cellular automata can be used to animate the static pixels. Hence, cellular automata can be used to produce interesting patterns and images in electrochromic displays, even with a rather small number of pixels. On the other hand, it is somehow laborious to design cellular automata rules to have a specific behaviour or to produce a specific pattern, unless the rules for that specific outcome are already known.

The use of pictorial entities as pixel elements provides a different approach to the presentation of visual information. The association of specific behaviour and interaction rules extended with cellular automata formulation to unique pictorial entities enables in turn an object oriented simulation of natural systems with inherent temporal and spatial dimensions. The approach presented is based on a multi-layer architecture of stackable electrochromic displays. It intends to provide a tangible way for experimenting with complex systems using intuitive transition rules and obvious pictorial entities. The real engineering challenges lie in the design and development of the necessary hardware.

6 Conclusions and Future Research

The research work undertaken in this thesis is based on the premise that computers should be invisible. Its use and presence should be so natural that they would become part of our surroundings and daily activities. Microcomputers would be integrated into everyday objects, augmenting a physical object's original use-value with a new set of digital functionalities.

The advances made in microelectronics and communications technology in the last two decades have pushed the technical vision of Ubiquitous Computing into the realm of the possible. Computers have become mobile and ever connected to the internet. Tablet computers or more clearly e-book readers are gradually overcoming the paradigm of the general-purpose personal computer in favour of small, specialised digital devices that are entering our lifestyle. However, we are not yet in the presence of a truly invisible computer. It is evident that smaller, faster and cheaper microelectronics as well as better interconnectivity between devices are a fundamental aspect of the vision of Ubiquitous Computing, but materials capable of levelling the integration of the physical and digital worlds are as well essential.

Printed Electronics is presented as a ground-breaking new type of electronics that are able to enhance the current state of Ubiquitous Computing. It promises the mass production of low-cost, lightweight, thin and flexible digital devices free from form factors. The idea behind is to use conventional printing technologies to produce passive and active electronic components in a wide array of substrates through the printing of electro-optical functional inks. The possibility of incorporating the concepts behind Printed Electronics with 3D printing is as well attractive. Complete digital devices and interactive objects could be created as a single part rather than as a case that encloses the circuit boards and individual electronic components. For instance, sensing and display elements could be directly embedded in the mechanical structure of the object being created instead of being assembled separately and incorporated afterwards.

The idea that all these technologies will be available to the general public is as well a compelling one. Nowadays, there are already numerous individuals that are taking advantage of the potential of digital fabrication technologies not only to create the things that they desire and need, rather than using what someone else thought they needed, but more importantly to solve specific problems that directly affects them. Indeed, Personal Fabrication is empowering individuals by giving them the control over technology and its development, whilst fulfilling their desires.

Making Printed Electronics technologies available to the general public can further pave the way for the development of a technological world shaped by its users and a user-driven reality of Ubiquitous Computing. It will encourage new ideas and products to come to life whilst giving end-users the ability to develop their own embedded digital devices. Likewise the internet ended up being shaped by its users and its utility adapted by each one of us, an open Internet of Things might as well be produced by all of us.

The realisation of the vision of Ubiquitous Computing is naturally dependent on the development of various technologies. For example, advances in wireless communications, energy storage, human-to-machine interfaces, sensors and digital display screens are of particular significance. In this thesis it was given a clear emphasis to the latest. Visual communication has always played an important role in human societies and nowadays, more than ever, is often used as a method of choice for conveying information.

In order to achieve a truly invisible computer, new ways to provide and present digital information have to be implemented. These must be able to present dynamic information and be easily incorporated into objects. Electrochromic displays have the potential to enable exciting novel forms of visual content than can be used to not only to entertain but also to inform and engage audiences to take action. From a point of view of creativity, it becomes possible to create hybrid contents that mix the traditional graphic print look and feel with features from the digital area, such as animated and readily updatable content. The effective use of such technology is nonetheless, subject to the strategic deployment of their unique features for a given situation. Different purposes can represent different architectures, with different complexities. Moreover, it should be taken into account that the technology is only an enabler. Careful attention must be given as well to the nature and presentation of the content which must be relevant and interesting to the audience at hand. As in traditional visual communication printed approaches, design considerations about the legibility, readability, typography, and imagery of the content must be taken into account in order to deliver an effective message.

These elements can be used to either emphasize important information or de-emphasize more trivial details. The possibility to add animations and movement to such elements means that not only new compositions and arrangements can be exploited but as well that all the elements have to be carefully integrated. In sum, a well-designed display can only be used to give a strong and clear message that successfully will draw attention, raise awareness, provide guidance, or simple entertain, if it delivers the appropriate content for the audience in question. Aesthetics and function have to be combined to provide a unique identity.

The proposed classification for electrochromic displays based on the type of architecture/content intended to illustrate the potential of these for presenting dynamic information beyond the traditional alphanumeric seven-segment arrangements and dot-matrix arrangements. It is composed by three main categories (type I to type III), with each category in its turn being divided in two sub-types (A and B). The examples developed for each type of architecture/content illustrate how these can be implemented as simple, task-specific applications following an end-user fabrication process. They all emphasize interaction and visual actuation, using the minimal hardware components. Likewise to what would be expected in a Personal Fabrication approach, all the materials and electronic components used to make the devices are commercially available online or in any electronics specialty store.

The complexity of the display architecture increases from type I to type III, likewise the ability to present dynamic visual content. Type A refers to the basic architecture/content, namely, for type I-A, the direct addressing of a pre-determined image; for a type II-A, the direct addressing of a set of geometric abstract segments that when combined form specific pre-determined visual elements; and for a type III-A, the matrix addressing of geometric abstract pixels capable of form bitmap images. Type B represents an enhancement in terms of content to the basic structure. In type I-B, the pre-determined image is segmented so its individual parts can be activated and animated separately; while in type II-B, the use of pictorial images (e.g. icons or glyphs) instead of abstract geometric picture elements not only enables new types of content but as well can provide a more visually stimulating approach and effective communication. Type III-B further explores the use of pictorial elements as pixels by associating to each pixel a specific meaning that influences the overall visual content of the display. It is highlighted its use as an interface for pictorial based simulations through extended cellular automata formulations.

Indeed, cellular automata are introduced here as a simple computation model capable of producing highly complex behaviour and visually interesting patterns. The spatial structure of cellular automata is equivalent to the matrix-based architecture of type III electrochromic displays, with the pixels of the display mirroring the individual cells of a cellular automaton. As such, the dynamic process of cellular automata can be explored to create dynamic visual content in electrochromic displays, though its use goes well beyond the creation of visually interesting patterns.

The ability to have pictorial elements as pixels in electrochromic displays instead of abstract geometric figures is further explored through the association of specific cellular automata-based behaviour and interaction rules to unique pictorial entities. The pictorial simulation approach followed attempts to provide an object oriented simulation of natural systems with inherent temporal and spatial dimensions. Its applicability in real-world, in the context of this thesis, was conceptualised through the development of a task-specific device based on a multi-layer architecture of stackable electrochromic displays. The main aim was to provide a simple and tangible way of experimenting with complex systems using intuitive and clear transition rules and pictorial entities. Ultimately, it is expected that it will lead to more ideas for applications and more sophisticated requirements for it. The main innovation of the proposed system relies in the integration of the different components. Even though the final device was not fully developed, due to current technology constrains, the technology concept and application were formulated, and experimentation and validation of the separate elements was done.

The aim of this thesis was to explore and demonstrate new possibilities for task-specific, low computation interactive devices. The various experiences and applications developed demonstrated that the technical universe explored (i.e. the exploit of printed circuits and electrochromic displays) offers new creative opportunities for inexpensive, flexible, visual communication digital devices. It also opens the doors for further research on the topic.

Electrochromic displays still require various improvements in ordered to achieve their expected potential. There is still a sizable gap between the theoretical performance of electrochromic displays and the actual working capabilities. Improvements in the resolution of the displays, contrast and lifespan are required. The limited variety of available colours and the slow response times are also observable drawbacks in certain applications. Further research is required as well in the development of adequate matrix-addressing control units and operating schemes. Passive matrix addressing schemes have the tendency to originate voltage leaks between neighbourhood pixels of active pixels, resulting in the colouration of undesired elements (the crosstalk effect). In turn, active-matrix addressing schemes are more complex and costly to manufacture that passive-matrix addressing electrochromic displays. Further investigation in terms of real world applications for electrochromic displays aimed at the presentation of dynamic visual information is also necessary. Most applications developed so far are either prototypes or technology showcases. It is necessary to deploy them in real world contexts and analyse how the public reacts to it.

From a Personal Fabrication ideal point of view, it would be interesting to explore and further develop fabrication technologies and processes that could make accessible Printed Electronics to the general public. Nowadays, Printed Electronics technologies are mainly available to specialised companies and R&D institutes. In an attempt to change this tendency, various companies launched recently crowdfunding campaigns to make available their Printed Electronics products to everyone. For example, Ynvisible successfully funded Printoo[30] on May 2004, an open-source printed electronic prototyping platform of paper-thin circuit boards and modules, and AgIC[31], named after Ag Inkjet Circuit, funded on April 2014, transforms home inkjet printers into Printed Electronic circuit board manufacturing equipment. Another interesting example is Circuit Stickers[32], a set of adhesive peel-and-stick electronics for crafting circuits that can be used in combination with conductive materials such as conductive paint or thread to build interactive projects without any complicated equipment or programming skills. All these examples illustrate solutions aimed at facilitating the fabrication process of electronic circuits whilst enabling electronics to be integrated in a range of non-traditional material. They also have the potential to be an effective education tool for the general public. More approaches of this nature would be more than welcome.

Last but not least, it would be interesting to further explore how the simplicity of cellular automata can be exploited not only to create interesting visual patterns in electrochromic displays but as well as a way to operate ubiquitous computing devices. The proposed prototype is a very specific example idealised for the simulation of dynamic systems that also needs to be further developed to bring the concept towards a higher technology readiness level. The basic technological components for such device are all identified but still need to be integrated with

30 http://www.printoo.pt
31 http://agic.cc/
32 http://chibitronics.com/

reasonably realistic supporting elements so they can be tested in a real world environment.

In sum, the main contributions of this research work can be listed as:

- Contextualization of the potential of Printed Electronics and Personal Fabrication in driving Ubiquitous Computing;

- Development of task-specific, visual information applications using direct addressing and passive-matrix addressing electrochromic displays and open source hardware;

- Systematization of visual content types in electrochromic displays;

- Reframing of the use of Pictorial Entities as a tangible way of experimenting with complex systems through the use of matrix addressing electrochromic displays.

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Annexes

Annex A - Electrochromic Materials

Electrochromic materials are known for having the property to change of colour when affected either by an electron-transfer process or by a sufficient electrochemical potential. In the following sub-sections, it is reviewed the most important classes of electrochromic materials for electrochromic displays: transition metal oxides; Prussian blue systems; viologens; conjugated conducting polymers; and metal coordination complexes (metallopolymers).

Transition metal oxides

Transition metal oxides form a series of compounds with a uniquely wide range of electronic properties. They have important applications as dielectrics, semiconductors and metals, and as materials for magnetic and optical uses (Cox, 1995). Several metal oxides films can be electrochemically switched from a colourless oxidised state to a non-stoichiometric[33] redox state which has an intense coloured reduced form. A good example is the tungsten trioxide (WO_3). Since its electrochromic properties were first reported in 1969 (Deb, 1969), it has been widely studied and today is still one of the most promising candidate for large-scale uses of electrochromic devices. Tungsten trioxide is transparent as a thin film, and upon electrochemical reduction has an intense blue colour. The detailed colouration mechanism is still controversial, but it is generally accepted that the injection and extraction of electrons and protons or univalent inert metal cations play a fundamental role (Mortimer, 2011; Rowley and Mortimer, 2002). The reaction can be written as shown in equation (A.1), where M^+ represents one of the following cations: H^+, Li^+, Na^+, or K^+; e^- designates the electrons; and $0 < x \leq 1$.

[33] Non-stoichiometric compounds are chemical compounds in which the numbers of atoms of the elements present cannot be represented by a ratio of well-defined natural numbers (see for instance, (Gusev et al., 2001)).

$$WO_3 + x(M^+ + e^-) \leftrightarrow M_xW^{VI}_{(1-x)}W^V_xO_3 \tag{A.1}$$

(transparent) (blue)

The oxides of molybdenum (MoO_3), vanadium (V_2O_5), niobium (Nb_2O_5), iridium ($Ir(OH)_3$), and nickel ($Ni(OH)_2$) are as well as of great electrochromic interest as they likewise present intense electrochromic colour changes. Equations (A.2) to (A.6) show the colouration mechanism of these elements.

Table A-1 summarises the colour states.

$$MoO_3 + x(M^+ + e^-) \leftrightarrow M_xMo^{VI}_{(1-x)}Mo^V_xO_3 \tag{A.2}$$

(transparent) (blue)

$$V_2O_5 + x(M^+ + e^-) \leftrightarrow M_xV_2O_5 \tag{A.3}$$

(yellow) (blue)

$$Nb_2O_5 + x(M^+ + e^-) \leftrightarrow M_xNb_2O_5 \tag{A.4}$$

(transparent) (blue)

$$Ir(OH)_3 + OH^- \leftrightarrow IrO_2 \cdot H_2O + H_2O + e^- \tag{A.5}$$

(transparent) (blue-black)

$$Ni(OH)_2 + OH^- \leftrightarrow NiO \cdot OH + H_2O + e^- \tag{A.6}$$

(pale green) (brown-black)

Table A-1: Colour states of relevant electrochromic transition metal oxides.

Transition metal oxides		**Colour**	
		Oxidised state	Reduced state
Tungsten Trioxide	WO_3	Transparent	Blue
Molybdenum Trioxide	MoO_3	Transparent	Blue
Vanadium Pentoxide	V_2O_5	Yellow	Blue-black
Niobium Pentoxide	Nb_2O_5	Transparent	Blue
Iridium Hydroxide	$Ir(OH)_3$	Blue-black	Transparent
Nickel(II) Hydroxide	$Ni(OH)_2$	Brown-black	Pale-green

In addition to the transition metal oxides listed in

Table A-1, the oxides of the following (transition) metals are as well electrochromic: cerium, chromium, cobalt, copper, iron, manganese, palladium, praseodymium, rhodium, ruthenium, tantalum and titanium. However, as their optical properties are not intense, they are usually more functional in electrochromic displays as an optically passive electroactive layer.

Prussian blue systems

Prussian blue (iron(III) hexacyanoferrate (II)) was the first artificially synthesised pigment (Ware, 1999). It was discovered unintentionally by Diesbach in 1704, and due to the limitations of existing blue pigments at the time, it rapidly attracted much attention. By the beginning of the nineteen century, Prussian blue add become a popular pigment, being used by artist in paints, by the papermaking industry as the principal colouring agent for blue toned papers, and as a constituent of several writing inks.

Prussian blue is the prototype of a number of polynuclear transition metal hexacyanometallates (Granqvist, 1995; Monk et al., 2007; Mortimer, 1997; Somani and Radhakrishnan, 2003). Their general formula is $M_b' \, [M''(CN)_6]_c$ where M' and M'' are transition metals with different oxidation numbers, and *b* and *c* are integral numbers. In these compounds, a given element exists in two oxidation states with the possibility of charge transfer between them. The chemical synthesis of Prussian blue is performed by the mixture of $[Fe^{III}(CN)_6]^{3-}$ or $[Fe^{II}(CN)_6]^{2-}$ and $Fe(II)$ or $Fe(III)$, respectively. The intense blue colour of the Prussian blue arises from the charge transfer transition between the mixed valence iron oxidation states (Robin, 1962).

The electrochromism of Prussian blue as a thin film was first reported in 1978 (Neff, 1978). It was observed at the time, that the electrochemical reaction of a Prussian blue thin film on a platinum foil electrode resulted on the anode being coloured bright blue, whereas the cathode was colourless. The electrode reaction evidently occurred in the film itself and corresponded to the reduction of Prussian blue to the colourless Prussian white, a species usually known as Everitt's salt (Equation (A.7)). Furthermore, it was possible to change rapidly between the blue and colourless states by switching the voltage potential.

$$\underset{\text{(blue)}}{[Fe^{III}Fe^{II}(CN)_6]^- + e^-} \leftrightarrow \underset{\text{(colourless)}}{[Fe^{II}Fe^{II}(CN)_6]^{2-}} \tag{A.7}$$

A second feature observed was related with the oxidation of the Prussian blue film at more anodic potentials, where it became green. In this case, the Prussian blue was oxidised to Prussian green, also known as Berlin green (Equation (A.8)). Again, the colour could be switched rapidly between blue and green.

$$3[Fe^{III}Fe^{II}(CN)_6]^- \leftrightarrow Fe_3^{III}[Fe^{III}(CN)_6]_2[Fe^{II}(CN)_6]^- + 2e^- \tag{A.8}$$

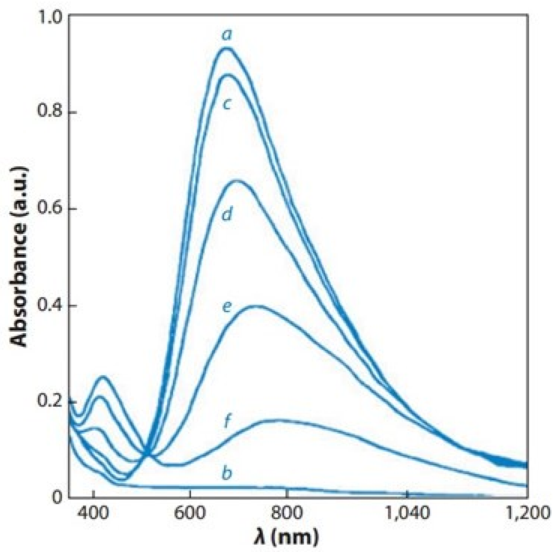
(blue) (green)

However, the transition of Prussian blue to Prussian green corresponded only to the partial electrochemical oxidation of the Prussian blue chromophore, as the complete oxidation results in a golden yellow Prussian yellow (Equation (A.9)).

$$[Fe^{III}Fe^{II}(CN)_6]^- \leftrightarrow [Fe^{III}Fe^{III}(CN)_6] + e^- \tag{A.9}$$

(blue) (yellow)

The electrochemical oxidation of Prussian blue hence presents a continuous mixed-valence composition between the yellow and blue states by the adjustment of the potential. By contrast, the reduction of Prussian blue to Prussian white is abrupt, involving the clean conversion of one element into the other at a critical potential (Mortimer and Rosseinsky, 1984). The spectra of the different forms of Prussian blue are shown in Figure A-1.



(a): +0.50V (blue)

(b): −0.20V (colourless)

(c): +0.80V (green)

(d): +0.85V (green)

(e): +0.90V (green)

(f): +1.20V (yellow)

Figure A-1: Spectra of iron hexacyanoferrate films on tin-doped indium oxide (ITO)/glass at various voltage potentials. Source: (Mortimer, 2011).

The first electrochromic display to use Prussian blue as electrochromic material was reported by Itaya *et al*. (Itaya et al., 1982). The seven-segment display used a single film of Prussian blue, sandwiched between two optically transparent electrodes. When an appropriate voltage potential was applied across the film, oxidation occurred near the positive electrode, producing Prussian brown, and reduction near the negative electrode, producing Prussian white. A Teflon spacer was used between the two electrodes.

Viologens

Viologens are salts that result from the diquaternisation of 4,4′-bipyridyl. The compounds are formally named as 1,1′-di-substituent-4,4′-bipyridilium if the two substituents *R′* and *R″* (see Figure A-2) at the nitrogen location are the same, and as 1-substitituent-1′-substituent′-4,4′-bipyridilium if they differ (Monk, 1998). The most extensively studied element from the viologens family is the methyl viologen (1,1′-di-methyl-4,4′-bipyridilium; or simply MV).

The electrochemical behaviour of viologens was first reported in 1933 (Michaelis and Hill, 1933). Initially they were investigated as redox indicators in biological studies due to possessing one of the lowest redox potentials of any organic system exhibiting a significant degree of reversibility. Subsequently they were the parent compounds for one of the most widely used herbicides in the world, the "paraquat" family (Bird and Kuhn, 1981).

Viologens exist in three main oxidation states (Figure A-2). The dicationic form (Figure A-2a) is the most stable of the three redox states, being colourless when pure. Reduction of the viologen dication results in the radical cation (Figure A-2b). Viologens radical cations are characterised for being strongly coloured, with high absorption coefficients, due to an intense intra-molecular charge transfer. The colours formed depend upon the choice of the nitrogen substituents (*R′* and *R″*). For instance, radical cations containing short alkyl groups are blue (blue-purple in concentrated solution), becoming crimson as the alkyl chain length increases (Rowley and Mortimer, 2002). Further reductive electron transfer yields the neutral specie (Figure A-2c). The intensity of the colour exhibited by bi-reduced viologens is low since no optical charge transfer or internal transition corresponding to visible wavelengths is available (Mortimer, 1997).

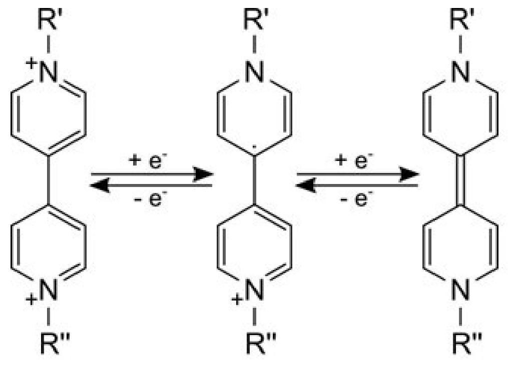


Figure A-2: The three common redox states of Viologens: a) dication, b) radical cation, and c) neutral species. Source: adapted from (Mortimer et al., 2006).

The first reduction step is highly reversible and can be cycled several times without significant side reaction. In contrast, the reduction to the fully reduced state (the neutral species) is less reversible, not only because the latter is frequently an insoluble species but also because is an uncharged one (Bird and Kuhn, 1981). Viologens can also be incorporated into polymers, the resulting materials retaining to a great extent the chemical and electrochemical properties of the viologen species.

The first application of viologens as electrochromic material in electrochromic displays was reported in 1973, by Schoot *et al*. (Schoot et al., 1973).

Conjugated conducting polymers

Conjugated conducting polymers (see for instance, (Chandrasekhar, 1999; Chilton and Goosey, 1995; Inzelt, 2012)) are organic polymers that exhibit high electrical conductivity. They are produced by either chemical or electrochemical polymerization of various aromatic molecules, such as pyrrole, thiophene, aniline, furan, carbazole, azulene or indole (Figure A-3).

The conductivity of the conjugated polymer is achieved through the process of “doping”. This essentially consists of the oxidation (p-doping), or in some cases reduction (n-doping), of the polymeric backbone by a number of simple anionic or cationic species (dopants) with the aim of creating charge carriers. Conjugated polymers possess a unique, extended π-conjugation structure, alternating simple and double bonds. The charge carriers formed upon doping confer a high mobility to the delocalised electrons[34] of the conjugated polymer, facilitating their movement along the polymeric backbone. The energy gap (bandgap) between the highest occupied π-electron band (valence band) and the lowest unoccupied band (conduction band) determines the electrical properties of the material. If the gap is large (e.g. 10 eV), electrons will be hard to excite into the conduction band, and thus the material will be an insulator at room temperature. In contrast, if the gap is small (e.g. 1.0 eV), then electrons may be excited from the valence band into the conduction band, and the material will present conductive properties. The energy gap of most conjugated conducting polymers is generally greater than 1 eV (Chandrasekhar, 1999).

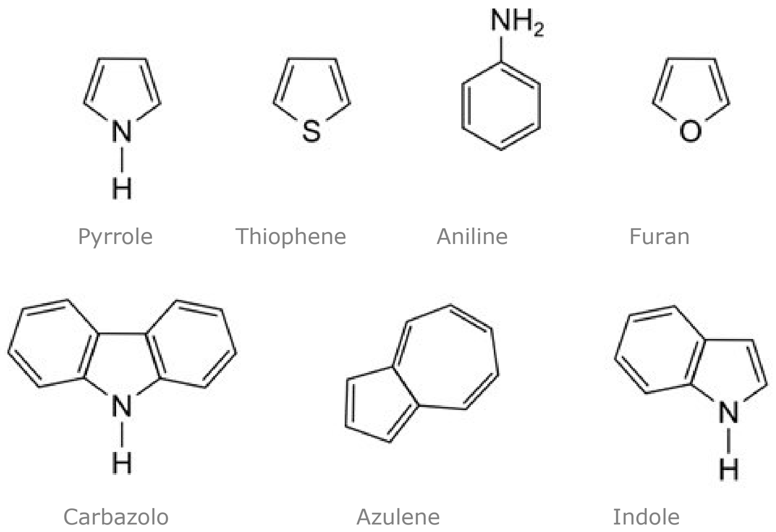


Figure A-3: Chemical structures of various aromatic compounds that can undertake chemical or electrochemical oxidation to produce conducting polymers. Source: adapted from (Rowley and Mortimer, 2002).

Typical anionic dopants include chloride (Cl^-), perchlorate (ClO_4^-), tetrafluoroborate (BF_4^-), hexafluorophosphate (PF_6^-), and polystyrene sulfonate ($[-CH_2CH(C_6H_4SO_3)-]_n^{n-}$); whereas a typical cationic dopant is sodium (Na^+) (Chandrasekhar, 1999). The main criteria in the choice of the dopant is its ability to oxidise or reduce the polymer without lowering its stability, and whether or not

[34] Delocalized electrons are not associated with a single atom or covalent bond. Hence, they are contained within an orbital that extends over several adjacent atoms.

they are capable of initiating side reactions that inhibit the polymers capability to conduct electricity.

The first chemical oxidative polymerization dates back to 1892 when Letheby (Letheby, 1862), while investigating two cases of fatal poisoning by nitrobenzol, and finding that this compound changed in the living stomach into aniline, decided to study into the chemical reactions of aniline. He observed that the electrolytic oxidation of a sulphatic solution of aniline resulted in a blue pigment (polyaniline), when the reduced form was colourless. However, the modern study of electric conduction in conjugated polymers only began in 1977 (see (Scott, 2010) for a thorough description), when Chian *et al.* reported the doping of polyacetylene (Chiang et al., 1977). Following this publication, numerous research groups began programs to understand better the properties of conjugated conducting polymers, and to search for new and better materials. Research papers on the topic became systematic. In less than a decade, most of the monomer building blocks that we know today had been identified and many procedures for polymeric synthesis had been documented. In 2000, the Nobel Committee recognised the crucial contributions of Heeger (Heeger, 2001), MacDiarmid (MacDiarmid, 2001) and Shirakawa (Shirakawa, 2001) in "[...] *the discovery and development of conductive polymers*" (Nobel Media AB, n.d.), awarding them the Nobel Prize in Chemistry.

All conjugated conducting polymers are potentially electrochromic in thin film form. They exhibit different colours whether they are in the oxidised (doped) or reduced (undoped) form. The colour change or contrast between the doped and undoped forms depends on the magnitude of the energy gap (bandgap) of the undoped form. Thin films with an energy gap greater than 3 eV (~400 nm) are colourless and transparent in the undoped form, while in the doped form their absorption spectrum is generally in the visible region. In turn, those with an energy gap equal to or less than 1.5 eV (~800 nm) are highly absorbing in the visible spectrum when in the undoped form but, after doping, the absorption in the visible spectrum is weak, being transferred to the near infrared. The polymers with the intermediate energy gaps are the ones more that present the most distinct optical changes throughout the visible spectrum (Mortimer et al., 2006).

Furthermore, conjugated conducting polymers can be tailored to induce other colour changes, i.e. they can be designed for a particular energy gap. This can be achieved by changing the composition of the polymers at the molecular level using a variety of synthetic strategies (see (Beaujuge and Reynolds, 2010; Stenger-Smith, 1998)) such as, varying the overall planarity of the polymer backbone, or by copolymerizing different monomers. The blending of different electroactive components as well as the creation of laminates and composites with other types of chromophores or insulating material are also suitable options. For example, the colours of thin films prepared from 3-methylthiophene-based oligomers are strongly dependent on the relative positions of methyl groups on the polymer backbone (Mastragostino et al., 1992, 1993). Hence, according to the position of the methyl groups, the films can either be pale blue, blue or violet in the oxidised form, and purple, yellow, red or orange in the reduced form.

These strategies are also frequently adopted to obtain polyelectrochromic devices, by combining electrochromic materials with different colour regions (Brotherston et al., 1999; Mudigonda et al., 1999). Composites between organic and inorganic materials have also been produced, not only with the aim of creating multicolour devices but also to decrease driving potentials. Examples reported in literature involve, for instance, polyaniline composite films combined with prussian blue (Duek et al., 1992, 1993; Morita, 1994).

In sum, the exploit of organic polymers as electrochromic materials presents several advantages with respect to inorganic electrochromic materials, not only in terms of flexibility, ease of process and low cost, but also with respect to both tailorability and efficiency of colouration (Carpi and De Rossi, 2006).

Box A-1: PEDOT

Poly(3,4-ethylenedioxythiophene)

Poly(3,4-ethylenedioxythiophene), often abbreviated as PEDT or PEDOT (Figure A-4), is a extensively studied and widely employed conjugated conducting polymer based on polythiophene. It was first developed by Bayer AG research laboratories (Heywang and Jonas, 1992; Jonas and Schrader, 1991), in the late eighties (1988), when researching for highly conducting polymers with low oxidation potentials, environmentally stable and soluble[35].

[35] See (Elschner et al., 2011) for an comprehensive historical overview of the discovery of PEDOT.

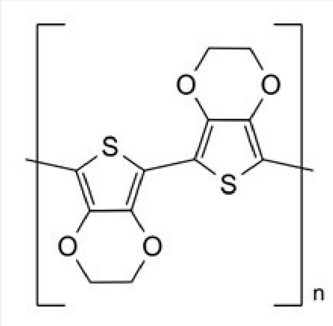


Figure A-4: PEDOT chemical structure.

PEDOT immediately draw the attention of the scientific community due to its unique properties. In addition to possessing a very high conductivity (~300 S/cm) and a high stability in the oxidised state, it also has an excellent transparency in thin, oxidised films (Dietrich et al., 1994; Heywang and Jonas, 1992; Jonas and Schrader, 1991). The energy gap of PEDOT is located at the transition between the visible and the near infrared regions of the spectrum (see Figure A-5).

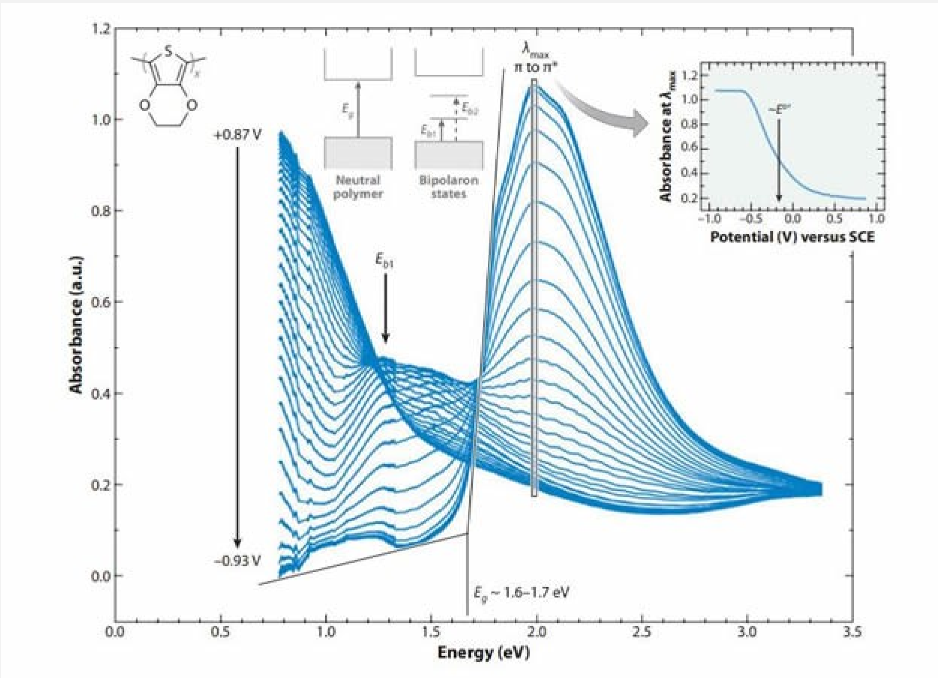


Figure A-5: Spectroelectrochemistry for a PEDOT film on tin-doped indium oxide (ITO).
Source: (Mortimer, 2011).

This low electronic bandgap (1.6-1.7 eV), which is the lowest in the polythiophene family, leads to very pronounced electrochromic properties. PEDOT is a strongly cathodically colouring material and much more transmissive to visible light in the doped and conducting state than it is in the reduced state (Pei et al., 1994). In this state, PEDOT exhibits a highly absorbing deep-blue colour while is almost transparent in the oxidise form, exhibiting a light-blue colour. Unfortunately, like most conducting polymers, PEDOT is insoluble and therefore difficult to process in thin film form. The solubility problem was eventually overcome by Bayer through the use of a water soluble polyelectrolyte, poly(styrene sulfonate) (commonly known as PSS), as the charge balancing dopant during the polymerization of PEDOT to yield PEDOT:PSS (Figure A-6). The result was a water soluble polyelectrolyte system with good film forming properties, high conductivity[36], high visible light transmissivity, and excellent stability (Groenendaal et al., 2000; Jonas et al., 1995). The lower price of PEDOT:PSS when compared to other alternatives also makes it a commercially attractive material.

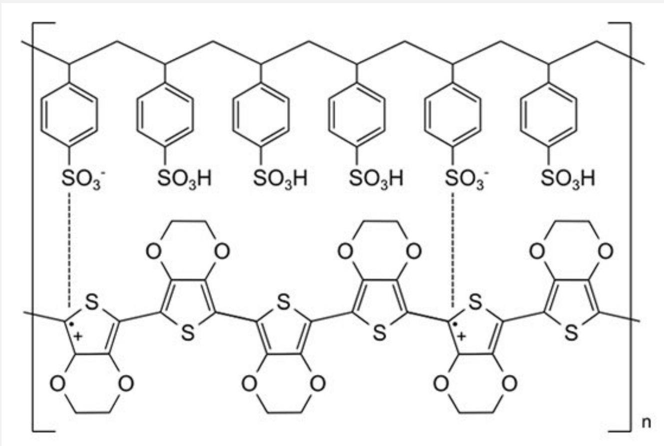


Figure A-6: Chemical structure of PEDOT:PSS. Source: adapted from (Monk et al., 2007).

The electrochromic colouration mechanism of PEDOT:PSS is show in Equation (A.10), where M^+ represents one of the following cations: H^+, Li^+, Na^+, or K^+; and e^- designates the electrons.

$$\underset{\text{(transparent light-blue)}}{PEDOT^+PSS^-} + M^+ + e^- \leftrightarrow \underset{\text{(deep blue)}}{PEDOT^0} + PSS^-M^+ \tag{A.10}$$

The physical properties of PEDOT:PSS make it possible for it to be selectively deposit, with a relative ease, into an assortment of substrates, whether they be rigid (e.g. glass) or flexible (e.g. paper). This is usually achieved by applying it as

[36] As an example, the commercially available solution produced by Heraeus with the trade name Clevios can have a conductivity of up to 1000 S/cm (Heraeus Precious Metals, 2012).

a water dispersion using standard coating or printing processes. Screen printing (see for instance, (McGarry and Tarr, 2008)), line patterning (see (Hohnholz et al., 2005)), and ink-jet printing (see (Tekin et al., 2008)) are the most commonly used manufacturing techniques. These methods not only reduce the number of fabrication steps but also eliminate the need for high-vacuum processing (Sankir, 2008).

The first description of a PEDOT based electrochromic device was reported by Gustafsson-Carlberg *et al.*, in 1995 (Gustafsson-Carlberg et al., 1995). The device was assembled when the authors were investigating the use of PEDOT and other conducting polymers of the polythiophene family in electrochromic applications (smart windows and displays). It consisted of a solid state electrochromic cell formed by one PEDOT layer on a tin-doped indium oxide (indium tin oxide, or simply ITO) coated glass electrode, and one vanadium oxide layer, also on an ITO coated glass electrode, separated by a solid polymer electrolyte. The use of PEDOT as the electrochromic material in solid state electrochemical cells demonstrated that it was possible to construct electrochromic devices with relatively small switching voltages (~1.5 V). In 2003, Argun *et al.* (Argun et al., 2003) reported the first truly all-polymer electrochromic device based on PEDOT:PSS. However, in this case, PEDOT:PSS was not used as the electrochromic layer but instead as the transparent electrode material, replacing the traditionally used ITO electrodes. As the electrochromic layer was used another cathodically colouring electrochromic polymer, and since the electroactive layer was also formed by an electrochromic polymer, the device was constructed using only organic and polymeric components. More recently, Andersson and co-workers (Andersson et al., 2007) combined an PEDOT:PSS based electrochemical transistor with an PEDOT:PSS based electrochromic display cell to form electrochemical smart pixels and build an all-organic active matrix addressed paper display (see also section 4.1.2.1.3). Here, PEDOT:PSS served both as the active material in the electrochemical smart pixels, as well as the conducting lines to route the updating signals of the integrated active-matrix display.

Metallopolymers

Metallopolymers (see for instance, (Maclanchlan, 2007; Wild et al., 2011)) are conjugated polymers that contain transition metal complexes. The great interest in these materials is related to their unique properties, which represent a combination of the physical, electronic and optical properties of the organic polymer with the physical, electronic and optical properties of the incorporated metal complex. Depending on the arrangement of the metal group relatively to the polymer backbone, metallopolymers can be divided into three types (Figure A-7) (Wolf, 2001):

- Type I metallopolymers have the metal group bind to the conjugated polymer backbone by a linker such as an alkyl group. The large connection distance between the polymer and the complexes results in no, or only weak, interaction between both. The polymer acts primarily as a conductive support and the electronic, optical, and chemical properties of the metal group are essentially the same as those of the unbind complex.

- In Type II metallopolymers, the metal is directly coupled to the conjugated polymer backbone making it easier for the polymer and metal group to affect each other's properties. Since π-conjugated backbones and many metal groups are redox active, the system might be able to be electrochemically tuned.

- Last, type III metallopolymers have the metal group directly incorporated into the conjugated polymer backbone. In this type of arrangement, strong electronic interactions between the organic bridge and metal group are possible. The properties of the conducting polymer are greatly influenced by the metal.

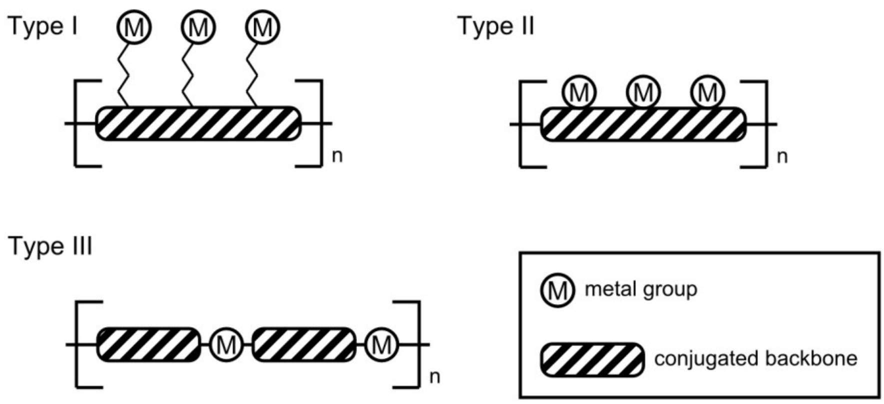


Figure A-7: Schematic representation of the three different types of structures of metallopolymers. Source: adapted from (Wolf, 2001).

The synthesis of metallopolymers can be done using a variety of methods (Wild et al., 2011). The most commonly used include condensation (Kingsborough and Swager, 2007), ring-opening metathesis (Buretea and Tilley, 1997) and electropolymerisation (Wolf, 2001). The successful free radical polymerization of vinyl ferrocene, reported in 1955, marks the first metal-containing polymer, poly(vinylferrocene) (Arimoto and Haven, 1955).

The use of metallopolymers as functional materials has been under investigation for the last decades, and a variety of potential applications are proposed (Holliday and Swager, 2005; Whittell and Manners, 2007; Whittell et al., 2011; Williams et al., 2007; Wolf, 2001). These include the use of metallopolymers as photoelectronic and electroluminescent devices; as photovoltaic materials; as electro-responsive sensors; as data storage devices; as molecular wires, switches, and antennae; and as macromolecular catalysts and artificial enzymes.

In terms of electrochromism, metallopolymers are attractive because of their intense colouration and ample redox reactivity. The chromophoric properties of metallopolymers usually emerge either from low-energy metal-to-ligand charge transfer (MLCT), intervalence charge transfer (IVCT), intraligand excitation, or related visible-region electronic transitions. Since all these transitions involve valence electrons, the chromophoric characteristics of the complex are altered or eliminated once it is oxidise or reduced (Monk et al., 2007; Zhang et al., 1992). Multicolour electrochromic changes can be induced by introducing two distinct types of metal ions into the polymer structure (see, (Higuchi, 2009; Higuchi et al., 2008)).

Good examples of metal coordination complexes with the potential for use in electrochromic devices include the $[M^{II}(bipy)_3]^{2+}$ series (Mortimer, 2011), where M^{II} corresponds to one of the following metal elements: iron (Fe), ruthenium (Ru), or osmium (Os); and $bipy$ to 2,2’-bipyridine. The choice of one metal complex over the other influences the colour of the metallopolymer film in the M^{II} redox state. Using iron as the metal complex confers a red colour to the thin film; while ruthenium gives it an orange colouration. In turn, osmium confers a green colour to the thin film. As an example, Zeng *et al.* (Zeng et al., 2008) reported on a Ru-phenolate based metallopolymer (metallopolymer incorporating a ruthenium metal centre) capable exhibiting rich electrochromic behaviour in the visible and near infrared regions of the electromagnetic spectrum, whereas in solution or in thin film form. The oxidation at 0.5 eV resulted in a change of colour from wine red to light green while potentials above 1.5 eV produced a change in the near infrared region.

Annex B - Types of Electrolytes

Electrolytes function as a source and sink of cations and anions as the various redox processes take place in an electrochromic displays. In the following sub-sections is briefly described the different classes of electrolytes commonly used in electrochromic displays.

Liquid electrolytes

Liquid electrolytes are characterised for having high ionic conductivities. The ionic transport is realised in a two-step process. First, the solvation and dissociation of the ionic compounds by the solvent molecules occurs, and second, the solvated ions migrate through the solvent medium. The excellent conductivity results from the combination of the ionic mobility and dissociation constant of the electrolyte system (Xu, 2004).

Solvent composition tailoring has been the main tool for manipulating electrolyte ion conductivity due to the availability of a vast number of candidate solvents (Xu, 2004). Since the advent of non-aqueous electrolytes, a wide spectrum of polar solvents has been investigated. The majority of them fall into either one of the following families: organic esters and ethers. Among these solvents, cyclic diesters of carbonic acid have undoubtedly attracted the main research attention. The most commonly used are propylene carbonate (PC), ethylene carbonate (EC) and γ-butyrolactone (γ-BL).

Ideally, liquid electrolyte solvents should: (1) be able to dissolve salts to a sufficient concentration; (2) have a low viscosity, so that facile ion transport can occur; (3) remain inert to all components of the electrochemical cell, especially the charged surfaces of the cathode and the anode; (4) remain liquid in a wide temperature range, i.e. its melting point should be low and its boiling point high; (5) be nontoxic; and (6) be economical (Xu, 2004).

Early electrochromic devices used liquid electrolytes as medium by excellence. For example, Deb's (Deb, 1969) novel electrochromic device used an aqueous solution of sulphuric acid (H_2SO_4) as electrolyte. Liquid acids are now rarely employed because of their tendency to degrade or dissolve electrochromic materials and because of fears that the device will leak (Mortimer, 1997). Another popular electrolyte solute was Lithium perchlorate ($LiClO_4$). However, the high oxidation state of chlorine (VII) in perchlorate makes it a strong oxidant, which readily reacts with most organic species in violent ways under certain conditions such as high temperature and high current charge (Jasinski and Carroll, 1970).

The main drawbacks in using liquid electrolytes are related to problems of leakage of the electrolyte solution, the weight gain from the solution, the low chemical stability, and homogeneity problems during the colouration process. A thickener, such as an acrylic polymer, poly(vinylbutyral), or colloidal silica, can be added to the liquid electrolyte solution to increase its viscosity and improve the safety of a electrochromic device should rupture occurs. Moreover, it helps minimize the effects of mass transport by natural convection. Clearly, as the addition of the polymer thickener is continued, the viscosity increases to the point where eventually the solution is self-supporting or free standing, and can be considered a physical gel (Byker, 2001).

Gel electrolytes

Gel electrolytes have a unique hybrid structure that consists of a polymer matrix swollen in a liquid medium (solvent). The liquid medium prevents the polymer matrix from collapsing into a compact mass whereas the polymer matrix, in turn, provides the microstructure to retain the liquid medium. The particularity of this arrangement is that the liquid medium, i.e. the solvent, is dissolved in the polymer matrix and not the other way around. The interactions between the polymer matrix and the liquid medium result in the electrolyte possessing simultaneously the cohesive properties of solid electrolytes and the diffusive transport properties of liquids electrolytes (Byker, 2001; Song et al., 1999).

Gel electrolytes are usually prepared by incorporating large quantities of a liquid plasticiser, alongside with the solvent(s), into the polymer matrix. The aim of the liquid plasticiser is to adjust the viscosity and improve the performance, namely the ionic conductivity, of the electrolyte. The excessive addition of the liquid plasticiser leads to the gel electrolyte presenting the same problems commonly associated to liquid electrolytes, as pointed out in the previous sub-section. Typical solvent plasticisers used are ethylene carbonate, propylene carbonate, dimethyl sulphoxide, dimethyl carbonate and diethyl carbonate.

Gel electrolytes are obtained as a result of either a chemical or a physical cross-linking process (Song et al., 1999). The chemical cross-link occurs when covalent bonds are created between the polymer chains by means of a chemical reaction to form a certain number of tie or junction points. In this case, as the number of junction points basically does not change upon the variation of the external conditions such as temperature, concentration, or stress, it leads to the formation of irreversible gels. Physical cross-link, on the contrary, occurs when the polymeric chains interact over a portion of their length or align in some regions to form small crystallites. In this case, "van der Waals" forces are responsible for joining the polymer chains and the solvent. Due to their weak nature, the connections can be broken by external conditions. The cross-linked polymer is typically formed by in-situ polymerization once the electrochromic device is filled with the solution containing the necessary polymer forming materials. Most gel electrolytes are based on polymer matrixes of polyacrylonitrile (PAN), poly(ethylene oxide) (PEO), poly(methyl methacrylate) (PMMA), poly(vinylchloride) (PVC), poly(vinyl pyrrolidone) (PVP), and poly(vinylidene fluoride) (PVDF).

The salts dissolved in the solution phase of a chemical or physical gel have ionic conductivities approaching those of liquid electrolytes. For example, gel electrolytes based on poly(methylmethacrylate) (PMMA) have room temperature conductivities of 10^{-3} S/cm (Agnihotry et al., 2000).

Solid electrolytes

Solid electrolytes generally have lower ionic conductivities than gel and liquid electrolytes. These lower conductivities arise from the slower ion diffusion through the solvated polymer matrix as a consequence of the higher viscosities. However, as electrochromic devices do not require high conductivity values due to the close proximity of the electrodes in the assembled electrochromic device, solid electrolytes that present ionic conductivities in the order of 10^{-4} S/cm at room temperature are adequate for use (Przyluski et al., 1993).

Solid electrolytes are obtained through the dissolution of salts in ion-coordinating macromolecules (the polymer matrix) liberated of any low weight solvent or additives (Bruce and Vincent, 1993). A wide variety of salts based on alkali-metal, alkaline-earth-metal, transition-metal and lanthanide ions can be used. The classic polymer host used is poly(ethylene oxide) (PEO) because of its solvation power and complexing ability to alkali metal ion (Liang and Kuo, 2004). Other commonly used polymer matrixes are polyacrylonitrile (PAN), poly(methyl methacrylate) (PMMA), poly(vinylchloride) (PVC), poly(vinyl pyrrolidone) (PVP), and poly(vinylidene fluoride) (PVDF), as already seen in gel electrolytes.

Solid electrolytes are typically formed by one of two methods. Thin films can either be fabricated by using solvent evaporation coating techniques, namely solvent casting, where basically the solvent is slowly removed from a homogeneous solution of polymer and salt; or, alternatively, by cryogenic grinding appropriate mixtures of polymer and salt and then subject the resulting powder to a modest heat treatment (Bruce and Vincent, 1993).

The advantage of using solid electrolytes in electrochromic devices is related to their mechanical and chemical stability. Furthermore, solid electrolytes have an excellent adhesive behaviour forming good interfacial contacts with the electrodes, and excellent elastomeric characteristics, being capable to relax elastically when under stress. In sum, they eliminate the problems associated with electrolyte evaporation and leakage, increase of hydrostatic pressure and device deformation, and undesired oxidation-reduction reactions with the electroactive materials. The need for a hermetic sealing is also eliminated.

Hybrid electrolytes

Hybrid electrolytes are organic–inorganic polymer electrolytes. The main synthetic route for the formation of these innovative systems is the sol–gel method (see (Brinker and Scherer, 1990)). The process consists of the formation of a gel system from a colloidal suspension of solid particles in a liquid, the sol. The sol is typically obtained through the hydrolysis and partial condensation of a precursor such as an inorganic metal salt or a metal organic compound (usually a metal alkoxide). The high versatility of the sol-gel method and the wide variety of precursors offers important advantages for the preparation of hybrid organic–inorganic electrolytes. It allows the incorporation of various organic polymers and molecules in the inorganic polymers, and thus the preparation of organic–inorganic hybrids that exhibit different physical and chemical properties depending on the organic phase and on the inorganic matrix (Orel et al., 2003). As a result, these hybrid materials combine the most important properties of their constituents, such as high transparency, low processing temperatures, sufficient thermal stability, and with high-performance yield, properties not found together in either material individually.

Based on the nature of the inner interfaces verified between the organic and inorganic phases, organic-inorganic hybrid materials can be divided into two distinct classes (Judeinstein and Sanchez, 1996): in class I (or type I), organic and inorganic compounds are held together only by weak bonds, namely hydrogen, van der Waals or ionic bonds; in class II (or type II), the two phases are linked together through strong chemical bonds, such as covalent bonds. Hybrid electrolytes should preferably be of type II. The ionic conductivity of hybrid electrolytes is only marginally smaller than that of liquid electrolytes.

Ionic liquid electrolytes

Ionic liquids are organic salts with a low melting point (below 100 ºC). They are, hence, typically liquid at room temperature. The most impressive feature of ionic liquids is the wide range of possible variations in their properties. In theory, ionic liquids can be designed to deliver almost any set of physical and chemical properties for almost any application in the chemical sciences (Freemantle, 2009). The tailoring of the physical, chemical and biological properties of ionic liquids is commonly done by changing the nature of the cations or anions; by introducing specific functionalities directly into the cations or anions, i.e. by changing their structure; or by mixing two or more simple ionic liquid (see (Freemantle, 2009; Seddon et al., 2000)). However, in practice, ionic liquids are usually selected for an application based on their properties rather than being specifically designed for it.

The unique properties that certain ionic liquids present has made them emerge as promising elements for electrolyte systems in electrochemical devices. In general, they have a high ionic conductivity (~10^{-2} S/cm) associated with excellent chemical and thermal stabilities. Furthermore, they feature low or negligible volatility, low flammability, and a wide electrochemical potential window. Two main strategies have been pursued by the scientific community in an attempt to translate the benefits of ionic liquids to polymer electrolytes. The first strategy involves the design of electrolyte systems composed of conventional polymer matrixes and ionic liquids; while the second consists of designing functional polymers presenting some of the characteristics of ionic liquids (Marcilla et al., 2006).

The implementation of ionic liquid based electrolytes in real electrochromic devices demonstrated that ionic liquids can indeed radically improve the performance, speed, cyclability and long term stability of electrochromic devices (Lu et al., 2002, 2003; Marcilla et al., 2006). The benefits of ionic liquids based electrolytes in electrochromic displays was first reported by Lu *et al.* (Lu et al., 2002).

Examples of ionic liquids suitable to be used in electrolytes for electrochromic devices are: ethyl ammonium nitrate ($[EtNH_3][NO_3]$), 1-butyl-3-methylimidazolium tetrafluoroborate ($[BMIM][BF_4]$) or 1-butyl-3-methylimidazolium hexafluoro-phosphate ($[BMIM][PF_6]$).

Annex C - Laser Cutting Parameters

The Epilog Mini 24 - Legend Elite Series is a versatile CO_2 laser system able to cut and engrave a wide variety of materials such as wood, acrylics, plastics, cork, leather, and rubber. It has a 610 x 305 mm work area, and is capable to cut material up to a maximum thickness of 140 mm. Table C-1 provides the cutting and engraving parameters for the materials used in the assembly of the electrochromic displays.

Table C-1: Epilog Mini 24 - Legend Elite Series cut and engraving parameters.

	Cut		Engraving	
	Power	Speed	Power	Speed
PET-ITO film	3%	10%	2%	20%
NITTO DENKO double-sided adhesive tape	10%	10%	5%	10%

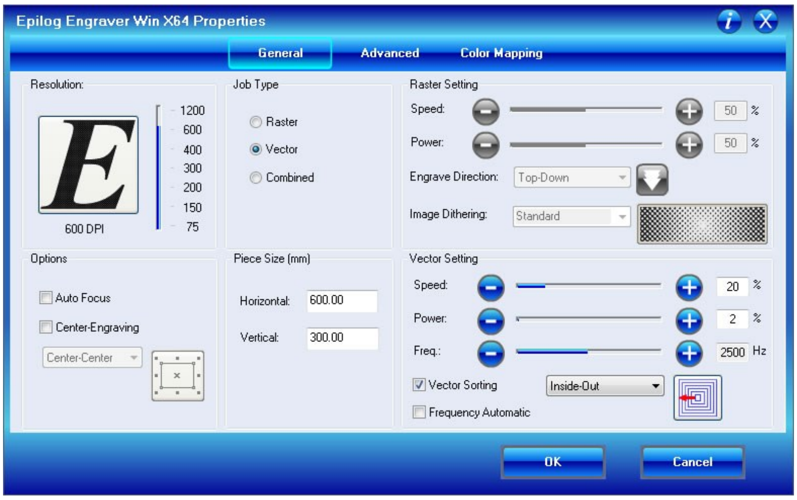


Figure C-1: Epilog Mini 24 - Legend Elite Series control panel.

Annex D - Direct Addressing ECDs Software

The software developed to operate direct addressing electrochromic displays was programmed using the Processing programming language and development environment. Figure D-1 illustrates the graphical user interface of the software. The top image depicts the initial screen where it is possible to choose the direct addressing arrangement of the electrochromic displays as well as select the computer COM port where the control unit is connect to. The following three images (bottom) illustrate the graphical user interface specific for each direct addressing arrangement (from left to right): the seven-segment numeral display, the four-segment die display, and a 4x4 matrix display.

Figure D-1: Graphical user interface of the control software used to interact with direct addressing electrochromic displays.

The diagram in Figure D-2 provides the source-code architecture of the control software developed for operating direct addressing electrochromic displays. Note that the diagram represents only is informal description of the most important characteristics of the architecture.

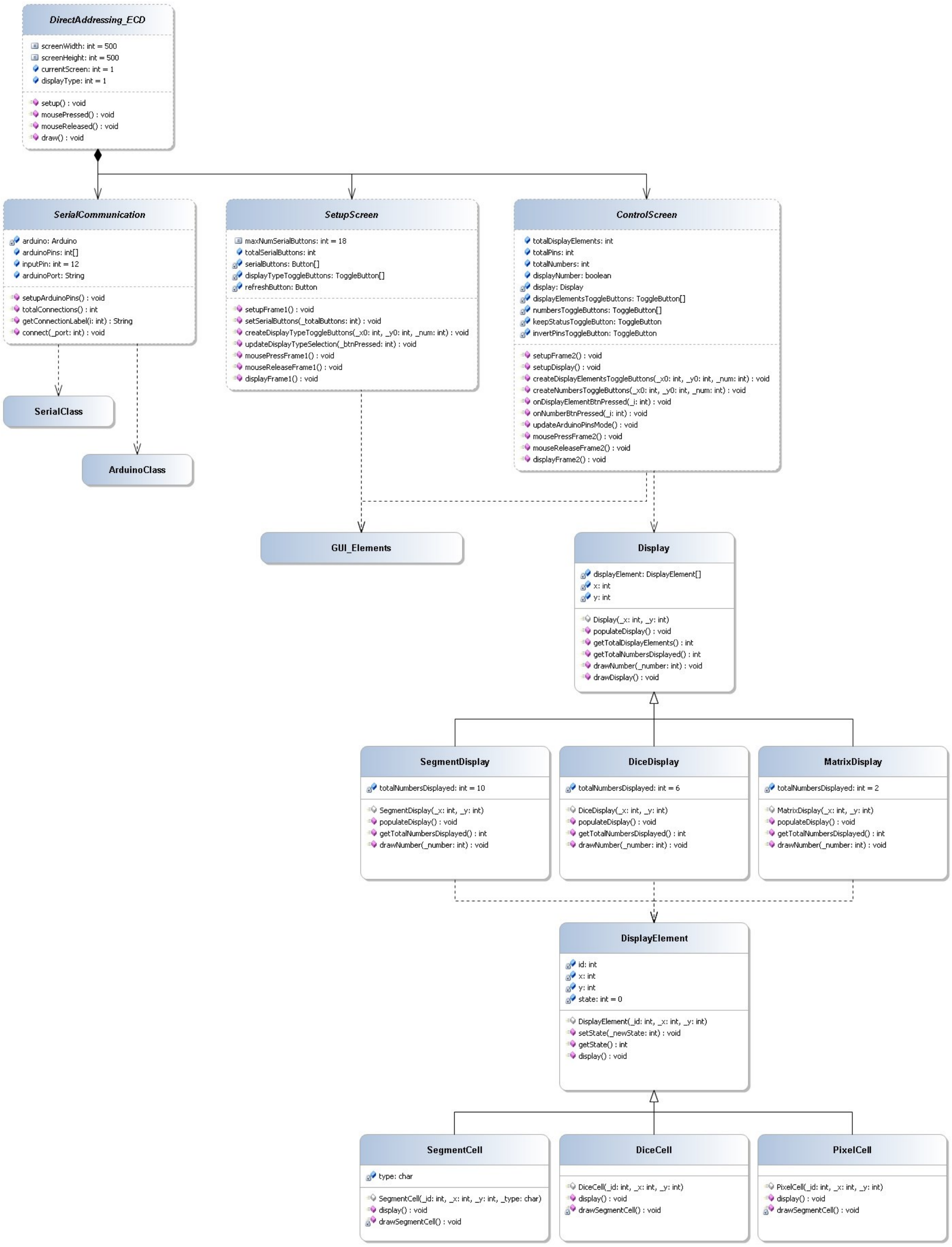


Figure D-2: Source-code architecture of the control software developed for operating direct addressing electrochromic displays.

Annex E - Passive-Matrix ECDs Software

The control software developed to operate passive-matrix addressing electrochromic displays was programmed using the Processing programming language and development environment. Figure E-1 shows the graphical user interface of the software. The top image presents the initial screen where it is possible to select the computer COM port to where the control unit is connect to, while the bottom image presents the graphical user interface for controlling a 4x4 passive-matrix display.

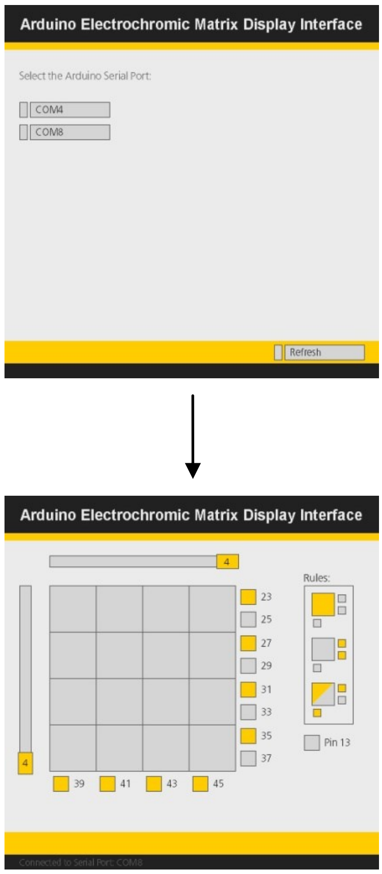


Figure E-1: Graphical user interface of the control software used to interact with passive-matrix electrochromic displays.

The diagram in Figure E-2 provides the source-code architecture of the control software developed for operating passive-matrix electrochromic displays. Note that the diagram represents only an informal description of the most important characteristics of the architecture.

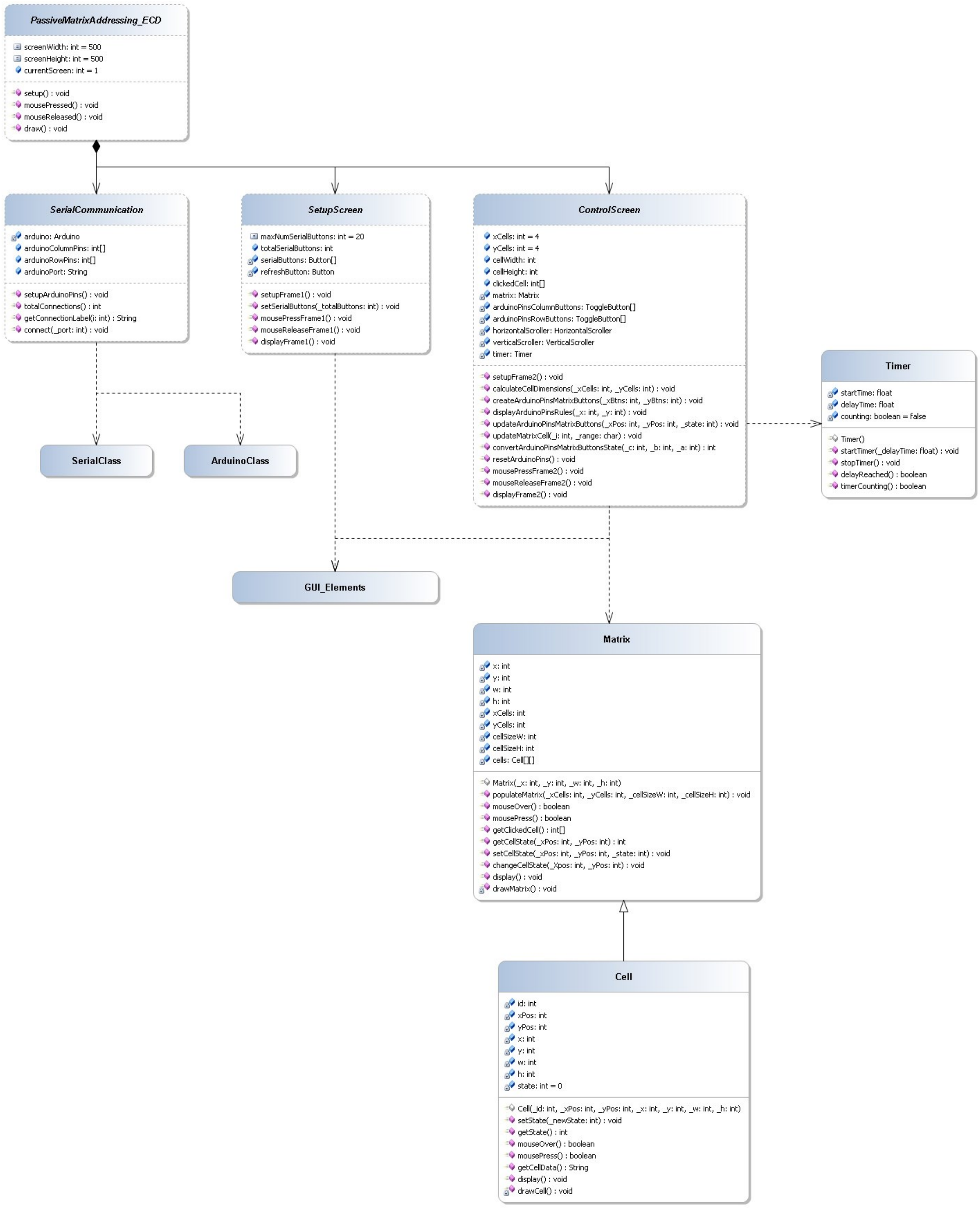


Figure E-2: Source-code architecture of the control software developed for operating passive-matrix electrochromic displays.

Annex F - Football Player Electrochromic Display Source-Code

```
/* *******************************************************
 * 5-Segment Football Player Animation Display
 * by Paulo Rosa
 * Upload Board: Arduino Duamilanove w/ATmega328
 * IDE: Arduino 1.0.4
 * NOTE: Makes use of PWM for 1.5V
 * *************************************************** */

// ATmega328 Pins Configuration
const int pinGround = 3;     // ground pin
const int pinSegA = 6;       // segment A PWM pin number
const int pinSegB = 5;       // segment B PWM pin number
const int pinSegC = 9;       // segment C PWM pin number
const int pinSegD = 10;      // segment D PWM pin number
const int pinSegE = 11;      // segment E PWM pin number
const int buttonPin = 1;     // button pin number

int buttonState = 0;
int buttonLastState = 0;
int buttonCounter = 0;

int time = 1000;

void setup ()
{
	pinMode (pinGround, OUTPUT);
	pinMode (pinSegA, OUTPUT);
	pinMode (pinSegB, OUTPUT);
	pinMode (pinSegC, OUTPUT);
	pinMode (pinSegD, OUTPUT);
	pinMode (pinSegE, OUTPUT);
	pinMode (buttonPin, INPUT);
}

void segmentsOFF ()
{
	analogWrite (pinGround, 0);
	analogWrite (pinSegA, 0);
	analogWrite (pinSegB, 0);
	analogWrite (pinSegC, 0);
	analogWrite (pinSegD, 0);
	analogWrite (pinSegE, 0);

	delay (time / 3);
}

void displaySegments (byte result)
{
	/* ******************************
	 * How to activate a segment?
	 * Segment Voltage == 0v
	 * Ground Voltage == 1.5v
	 * **************************** */

	analogWrite (pinGround, 76);

```

```
    /* ******************************************************************************
     * Result must come in the form of a binary value where 1 = ON, 0 = OFF
     * | Seg A    | Seg B    | Seg C    | Seg D    | Seg E    |
     * | B00001 | B00010 | B00100 | B01000 | B10000 |
     * *************************************************************************** */

    analogWrite (pinSegA, bitRead (result, 0) == 0 ? 76 : 0);
    analogWrite (pinSegB, bitRead (result, 1) == 0 ? 76 : 0);
    analogWrite (pinSegC, bitRead (result, 2) == 0 ? 76 : 0);
    analogWrite (pinSegD, bitRead (result, 3) == 0 ? 76 : 0);
    analogWrite (pinSegE, bitRead (result, 4) == 0 ? 76 : 0);
}

void testSegments ()
{
    segmentsOFF ();
    displaySegments (B00001);
    delay (time * 5);

    segmentsOFF ();
    displaySegments (B00010);
    delay (time * 5);

    segmentsOFF ();
    displaySegments (B00100);
    delay (time * 5);

    segmentsOFF ();
    displaySegments (B01000);
    delay (time * 5);

    segmentsOFF ();
    displaySegments (B10000);
    delay (time * 5);

    segmentsOFF ();
    displaySegments (B10101);
    delay (time * 5);

    segmentsOFF ();
    delay (time * 5);
}

void displayFootballPlayerSequence (int state)
{
  switch (state)
  {
    case 1:
     displaySegments (B01101);
      break;
    case 2:
      displaySegments (B01110);
      break;
    case 3:
      displaySegments (B11010);
      break;
    default:
      displaySegments (B00000);
  }
}

void loop()
{
  // read the state of the pushbutton value:
  buttonState = digitalRead (buttonPin);

  if (buttonState != buttonLastState)
  {
    if (buttonState == HIGH)
    {
      buttonCounter ++;

      segmentsOFF ();
      displayFootballPlayerSequence (buttonCounter);
      delay (time / 2);

```

```
      if (buttonCounter == 4)
        buttonCounter = 0;
    }
  }

  buttonLastState = buttonState;
}
// END
```

Annex G - Digital Dice Electrochromic Display Source-Code

```
/* ********************************************************
  * 4-Segments DIGITAL DICE Display
  * by Paulo Rosa
  * Upload Board: ATtiny85 (internal 8MHz clock)
  * IDE: Arduino 1.0.4
  * NOTE: Requires a 1.5V Power Source
  *
  * DESCRIPTION: Random numbers are generated every
  * time a button is pressed
  * ***************************************************** */

// Pin numbers:
const int buttonPin = 0;    // pin number of the TiltSensor
const int pinSegA =  4;      // pin number of the segment A
const int pinSegB =  3;      // pin number of the segment B
const int pinSegC =  1;      // pin number of the segment C
const int pinSegD =  2;      // pin number of the segment D

int buttonState = 0;
int buttonLastState = 0;
int randNumber;

void setup()
{
  pinMode (buttonPin, INPUT);
  pinMode (pinSegA, OUTPUT);
  pinMode (pinSegB, OUTPUT);
  pinMode (pinSegC, OUTPUT);
  pinMode (pinSegD, OUTPUT);
}

void setPinState (int pin, int state)
{
  // Activate de pin with the inverted result
  digitalWrite (pin, state == HIGH ? LOW : HIGH);
}

void clearResult ()
{
  setPinState (pinSegA, LOW);
  setPinState (pinSegB, LOW);
  setPinState (pinSegC, LOW);
  setPinState (pinSegD, LOW);
}

/* ****************************
  * #1 = Segment B
  * #2 = Segment A
  * #3 = Segment B + D
  * #4 = Segment A + D
  * #5 = Segment A + B + D
  * #6 = Segment A + C + D
  * *************************** */

void displayResult (int result)
{
```

```
  switch (result)
  {
    case 1:
      setPinState (pinSegB, HIGH);
      break;

    case 2:
      setPinState (pinSegA, HIGH);
      break;

    case 3:
      setPinState (pinSegB, HIGH);
      setPinState (pinSegD, HIGH);
      break;

    case 4:
      setPinState (pinSegA, HIGH);
      setPinState (pinSegD, HIGH);
      break;

    case 5:
      setPinState (pinSegA, HIGH);
      setPinState (pinSegB, HIGH);
      setPinState (pinSegD, HIGH);
      break;

    case 6:
      setPinState (pinSegA, HIGH);
      setPinState (pinSegC, HIGH);
      setPinState (pinSegD, HIGH);
      break;
  }
}

void loop()
{
  // read the state of the pushbutton value:
  buttonState = digitalRead (buttonPin);

  if (buttonState != buttonLastState)
  {
    if (buttonState == HIGH)
    {
       clearResult ();
       randNumber = random (1, 7);
    }

    displayResult (randNumber);

    //digitalWrite (ledPinA, randNumber == 1 ? HIGH : LOW);
    //digitalWrite (ledPinB, randNumber == 2 ? HIGH : LOW);
    //digitalWrite (ledPinC, randNumber == 3 ? HIGH : LOW);
    //digitalWrite (ledPinD, randNumber == 4 ? HIGH : LOW);
  }

  buttonLastState = buttonState;
}
// END
```

Annex H - Waste Reminder Electrochromic Display Source-Code

```
/* ******************************************************
 * 3-Segment Waste Reminder Display
 * by Paulo Rosa
 * Upload Board: ATtiny85 (internal 8MHz clock)
 * IDE: Arduino 1.0.4
 * NOTE: Requires a 1.5V Power Source
 *
 * DESCRIPTION: Icons are highlighted following a
 * pre-determined time schedule
 * *************************************************** */

#include <Time.h>
#include <TimeAlarms.h>

// Pin numbers:
const int pinSegA =  4;     // pin number of the segment A
const int pinSegB =  3;     // pin number of the segment B
const int pinSegC =  2;     // pin number of the segment C
const int pinGround = 0;    // ground pin

void setup()
{
    pinMode (pinSegA, OUTPUT);
    pinMode (pinSegB, OUTPUT);
    pinMode (pinSegC, OUTPUT);
    pinMode (pinGround, OUTPUT);
}

void setPinState (int pin, int state)
{
    digitalWrite (pin, state);
}

void activateSegments (byte result)
{
    /* ******************************
     * How to activate a segment?
     * Segment Voltage == 0v
     * Ground Voltage == 1.5v
     * **************************** */

    setPinState (pinGround, HIGH);

    // Activate de pin with the inverted result
    setPinState (pinSegA, bitRead (result, 0) == LOW ? HIGH : LOW);
    setPinState (pinSegB, bitRead (result, 1) == LOW ? HIGH : LOW);
    setPinState (pinSegC, bitRead (result, 2) == LOW ? HIGH : LOW);
}

void reset ()
{
    activateSegments (B000);
}

void wasteAlarm()
{
```

```
    activateSegments (B001);
}

void paperAlarm()
{
    activateSegments (B010);
}

void plasticAlarm()
{
    activateSegments (B100);
}

void loop()
{
    reset ();

    Alarm.alarmRepeat(dowTuesday, 19,30,0, wasteAlarm);
    Alarm.alarmRepeat(dowTuesday, 19,30,0, paperAlarm);

    Alarm.alarmRepeat(dowThursday, 19,30,0, wasteAlarm);
    Alarm.alarmRepeat(dowThursday, 19,30,0, plasticAlarm);
}
// END
```

Annex I - Pictorial Simulation System: Code Architecture

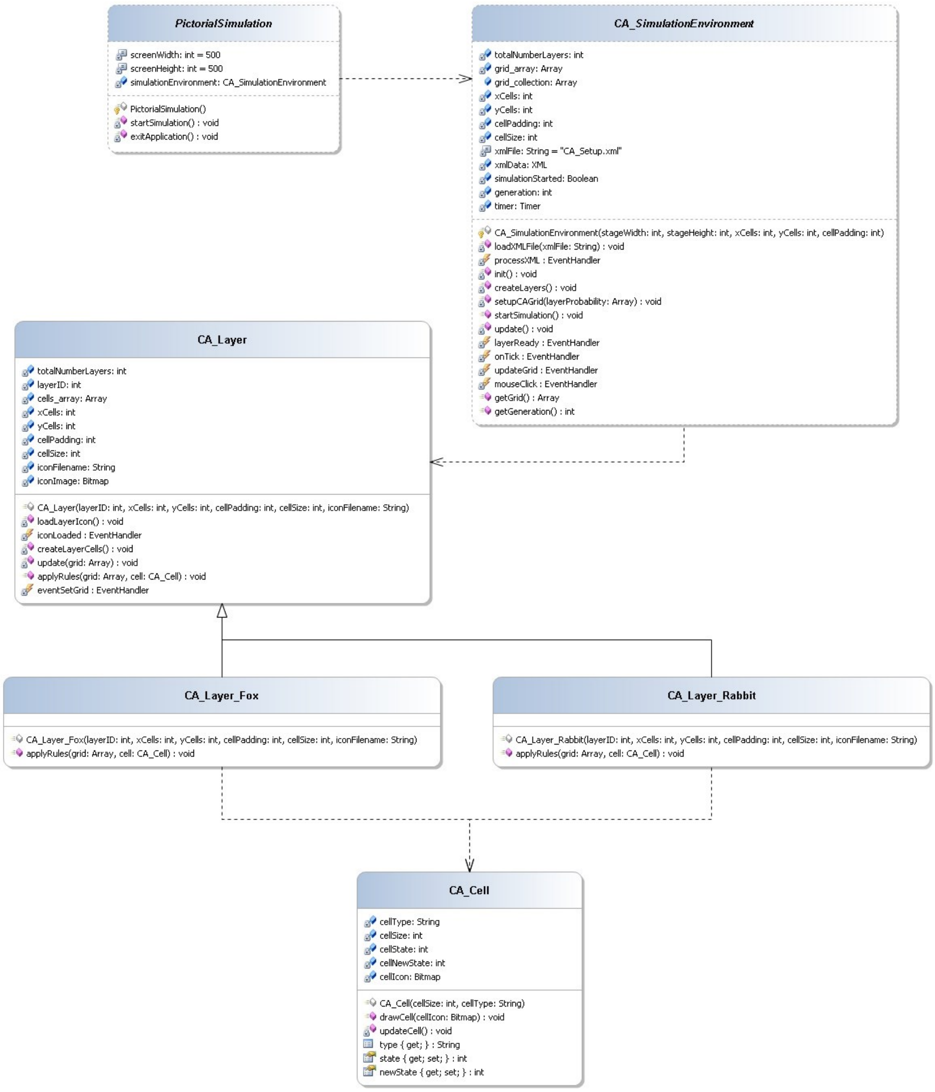


Figure I-1: Source-code architecture of the multi-layer pictorial simulation system.